Ultrafast Heating Experiments and Diagnostics

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Warm materials are dynamic

- energy deposition of ~ one quanta per unit cell drives structural and other property changes that can be probed by time-resolved, x-ray scattering

- a variety of quanta can be utilized to “heat” materials
  - THz to far-IR directly drives phonon modes
  - near-IR excites electrons from valence to conduction bands
    - with adjustable excess-energy that rapidly couples to other modes
  - optical to uv excites electronic transitions and charge transfer states
  - soft x-rays couple core levels to valence states
  - hard x-rays penetrate and excite larger volumes

- coupling of excitation to various modes is defined by the time-scale
  - times << picoseconds can involve non-thermal processes
    - photochemistry, electron re-scattering
  - times >> picoseconds involve thermal processes
    - mode diffusion, ablation
  - consider… relevant scale length <u>divided by</u> relevant velocity

Warm materials are studied by pump-probe techniques at a variety of facilities

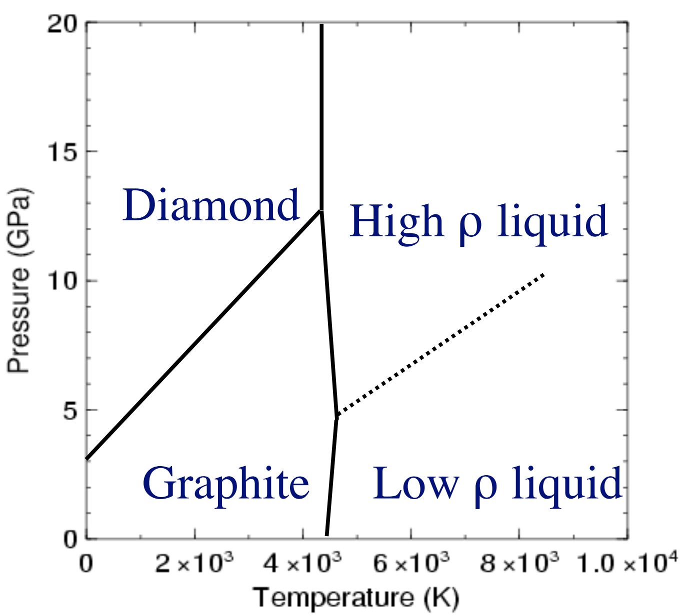
- <u>small-scale laboratories</u> provide intense, short-pulse lasers to create and probe warm (high-energy-density) materials
  - probes include plasma x-ray sources, high-harmonic sources

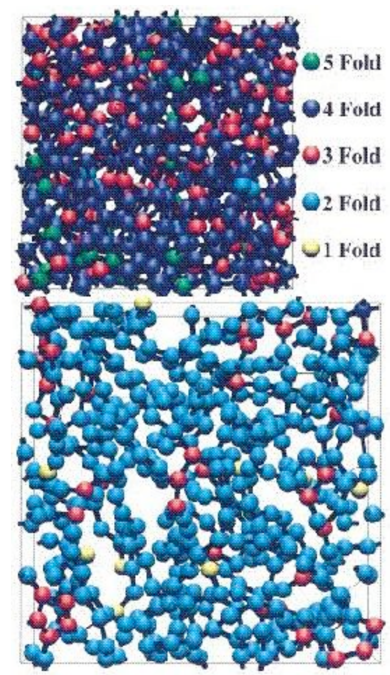
- <u>intermediate-scale facilities</u> include petawatt lasers, pulsed particle beams, x-ray synchrotrons,free-electron lasers, etc., and are widely accessible

- <u>large-scale facilities</u> allow large volume studies to extreme high-energy-density conditions, but have limited access
  - NIF megajoule laser, Vulcan PW laser, OMEGA kJ laser, pulsed power

An example of HEDS science: liquid carbon

Molecular dynamics calculations predict:
high density liquid
- mainly sp^3 coordinated
and low density liquid:
- mainly sp coordinated

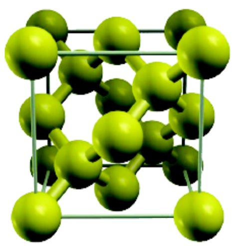




Glosli, et al, PRL *82*, 4659 (1999)

Phases of Carbon

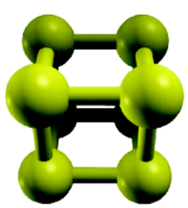
The goal is to study these phases
under extreme conditions,
liquid phases and melting lines



Diamond

BC8 (P > 1100 GPa)

- Body Centered Cubic with 8 atom basis
- Theoretical phase proposed in analogy with Si
- Semi-metallic, not yet found experimentally

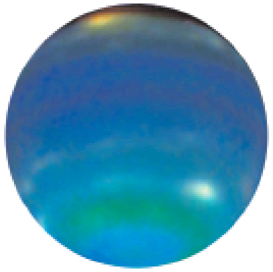


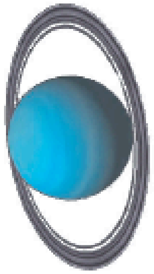
Cubic

- Metallic, not yet found experimentally

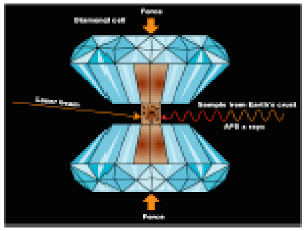
Liquid carbon

- Astrophysics and Planetary Science: State of Carbon in Giant Planets



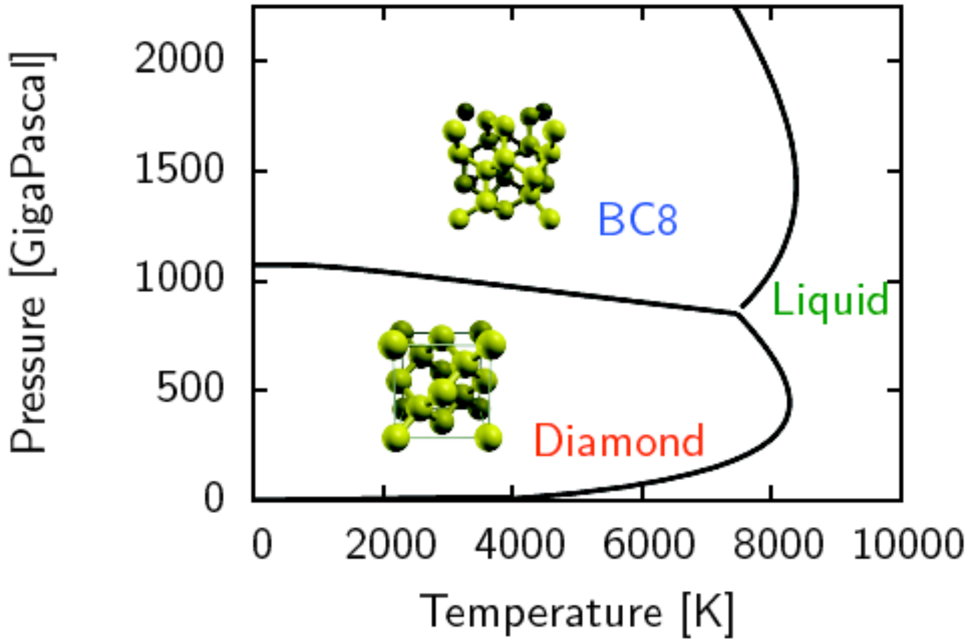


- High pressure research: Theoretical limit of diamond based technology



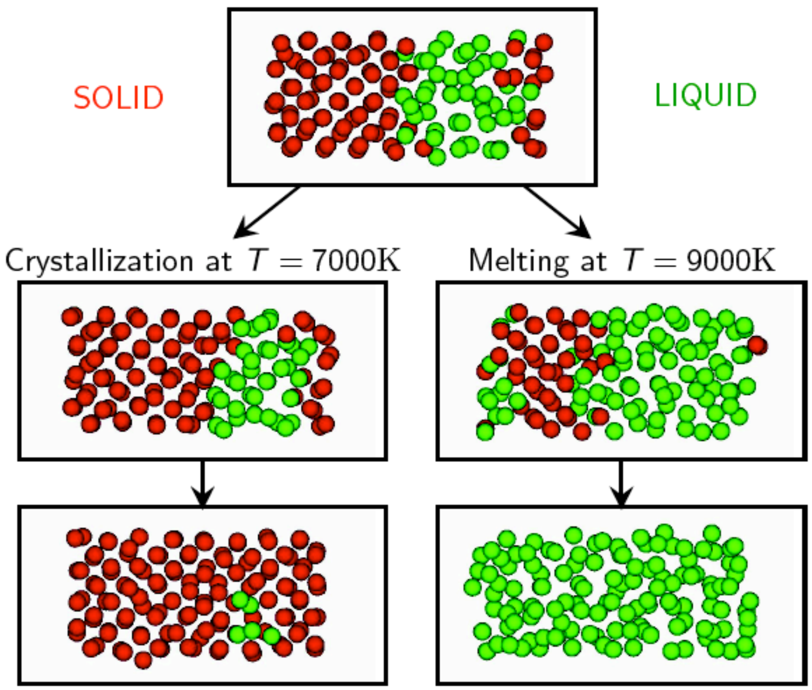
- Technological: Capsules for *Inertially Confined Fusion* (Carbon as an ablator material)

Phase Diagram of Carbon



- Predicted maximums in melting lines
- Triple point
- Negative slope of Diamond-BC8 transition
- Experimentally verified negative melting slope
  - *(P > 500 GPa) by Shock experiments (Eggert et al. 2007)*
- **Correa et al. PNAS 103(5) (2006)**

Melting lines obtained by the two-phase simulation method

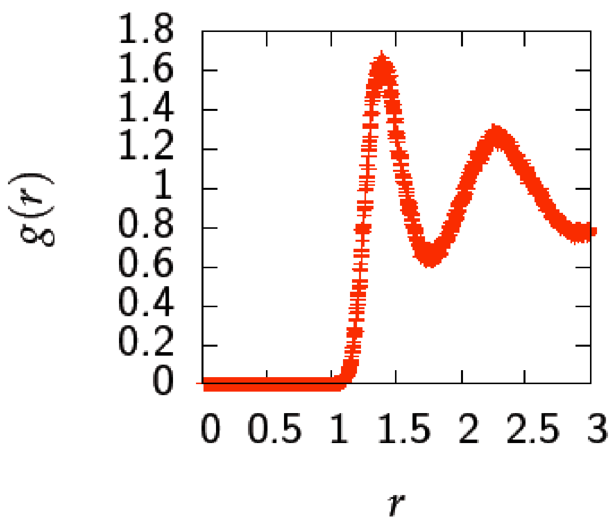


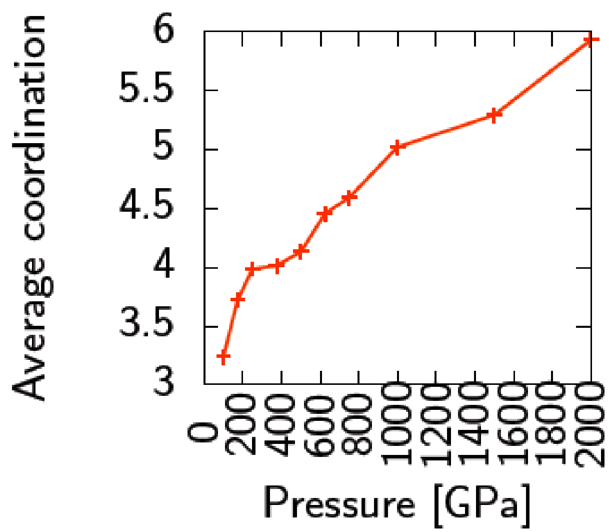
- Even with the most realistic molecular dynamic simulation, melting lines are not trivial to obtain
- Density Functional Molecular Dynamics on 128 carbon atoms
- Quantum mechanical electrons and Classical Ions
- Ab initio (no *fitted* parameters)
- Solid and Liquid initially present in same simulation
- Interface evolves at a given *P and T.*
- *Most stable phase grows*
- *Melting line is bracketed recursively*

Correa et al. PNAS 103(5) (2006)

Predicting absorption spectra

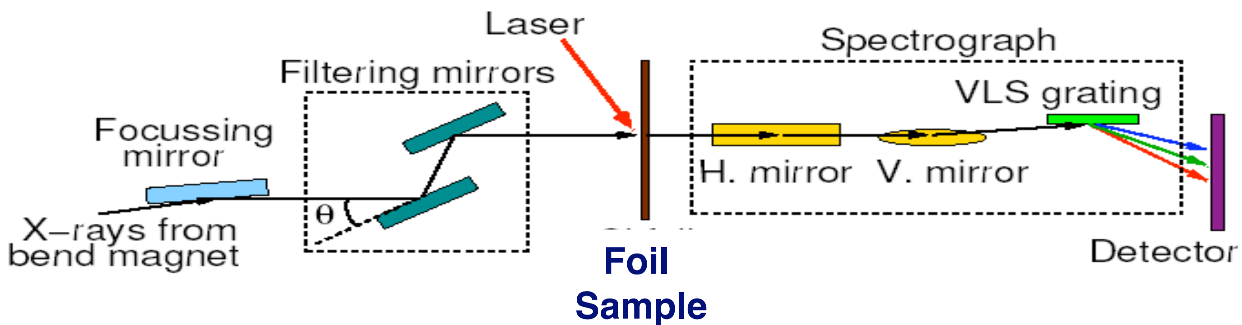
Quantum Molecular Dynamics does a great job in terms of predicting structural properties:

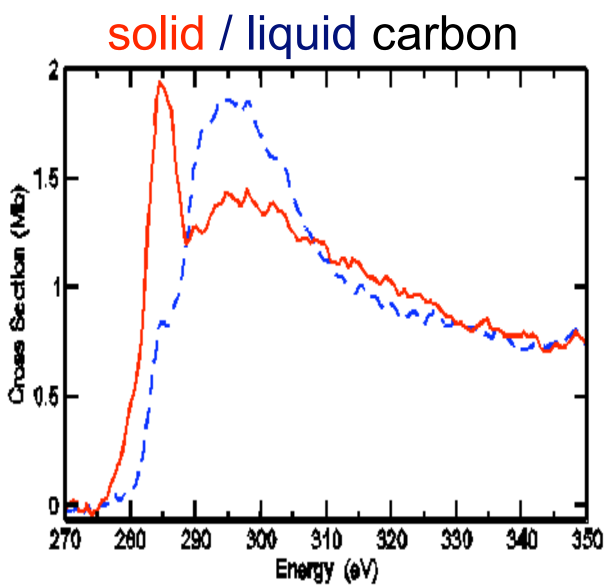


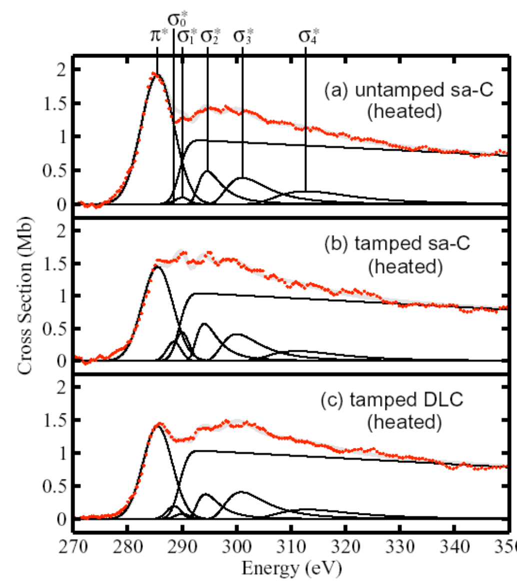


but we need to go further...
Can we reproduce experimental results and predict specific results for new conditions?

High-energy-density carbon has been probed by x-ray absorption (near and extended edge)



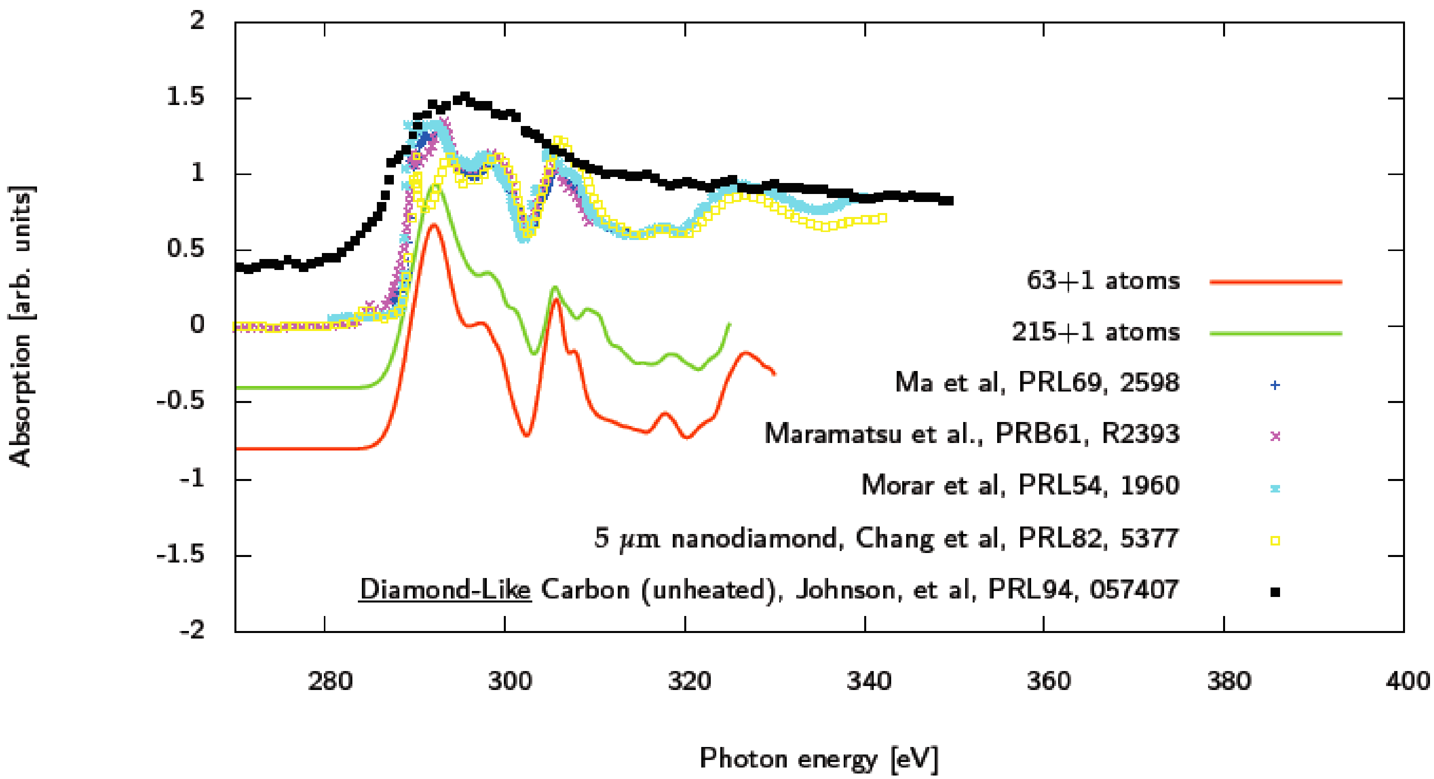




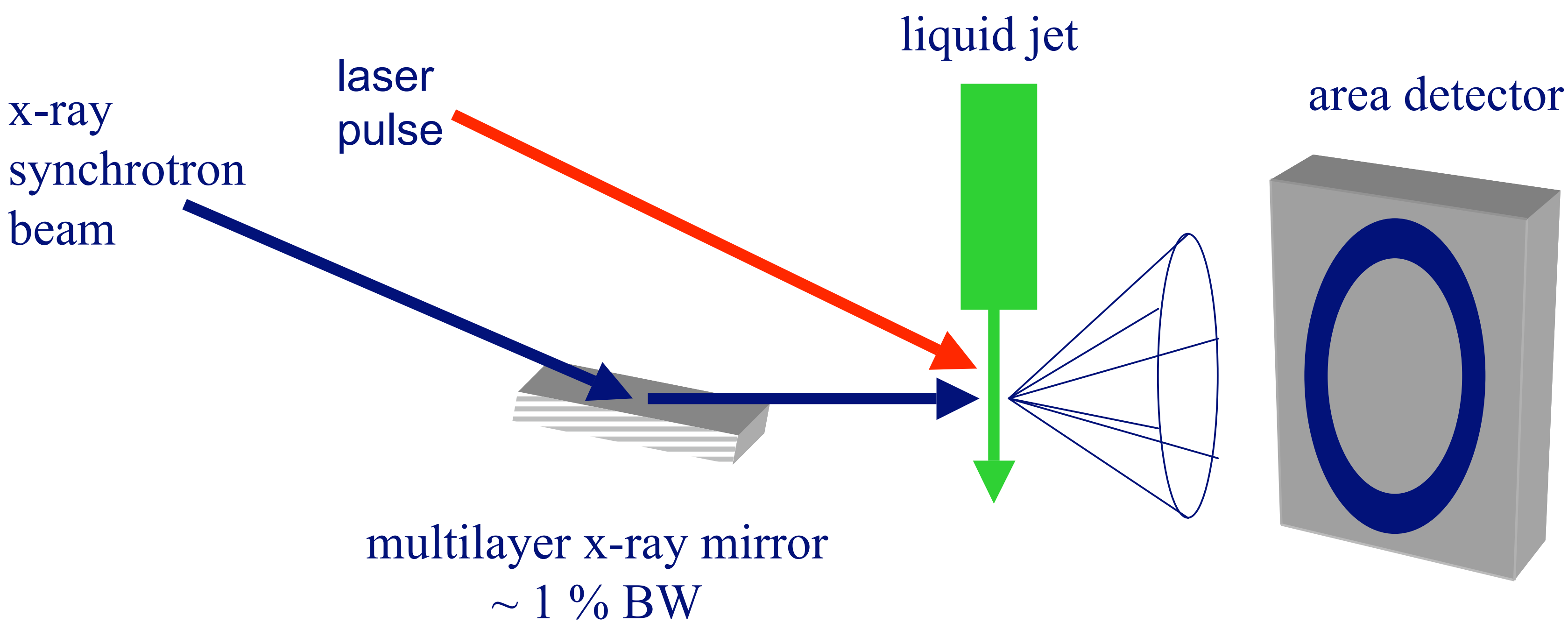
supports calculations indicating that the low-density phase of liquid carbon is predominately sp-bonded

S. Johnson, *et al*
Silicon: PRL **91**, 157403 (2003)
Carbon: PRL **94**, 057407 (2005)

Predicting high-T absorption spectra



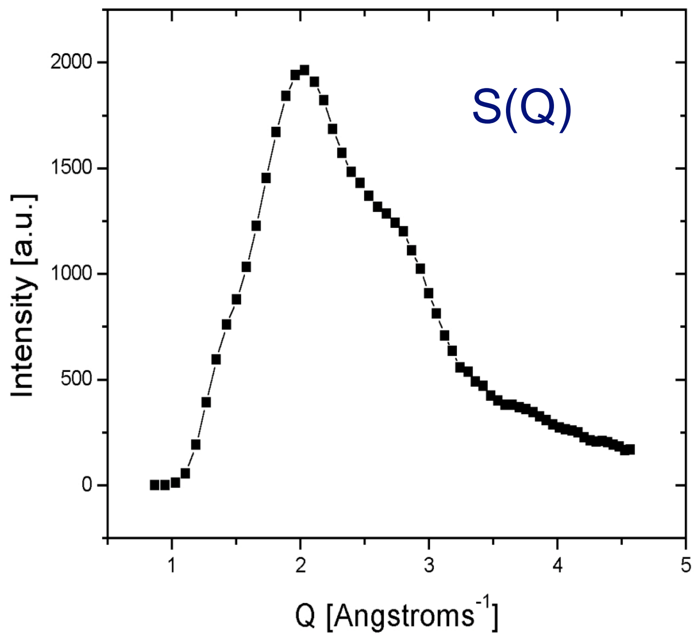
Perturbed liquid state structure and dynamics can be probed by x-ray scattering (small and wide angle)



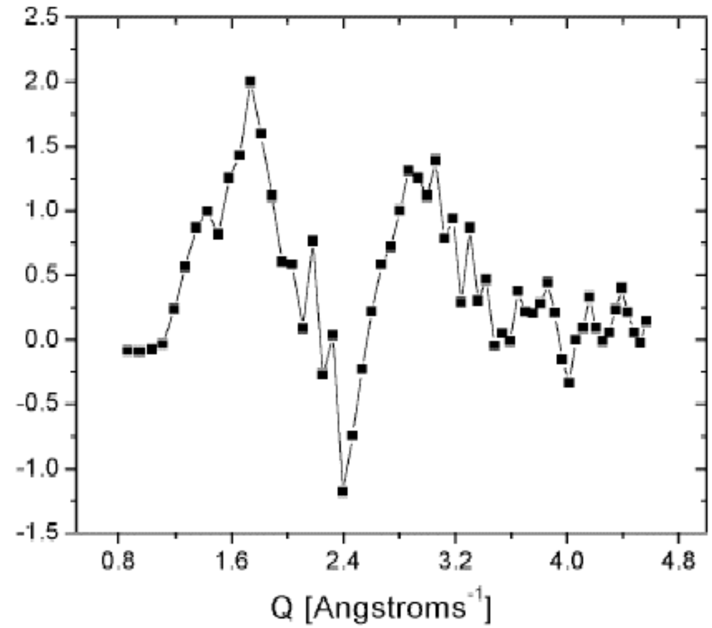
A. Lindenberg

Time-resolved structural changes in H_2O are seen upon charge injection

Static scattering signal



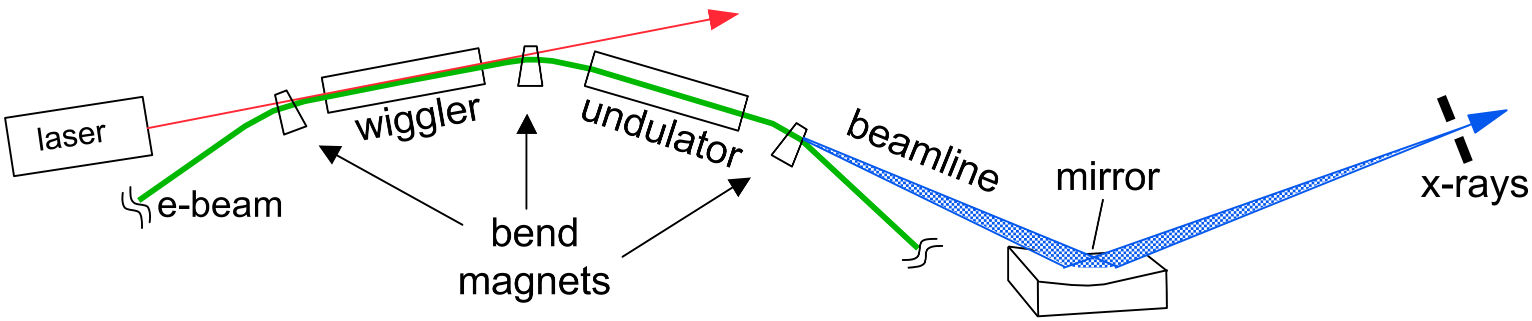
Difference signal at 100 ps following charge injection

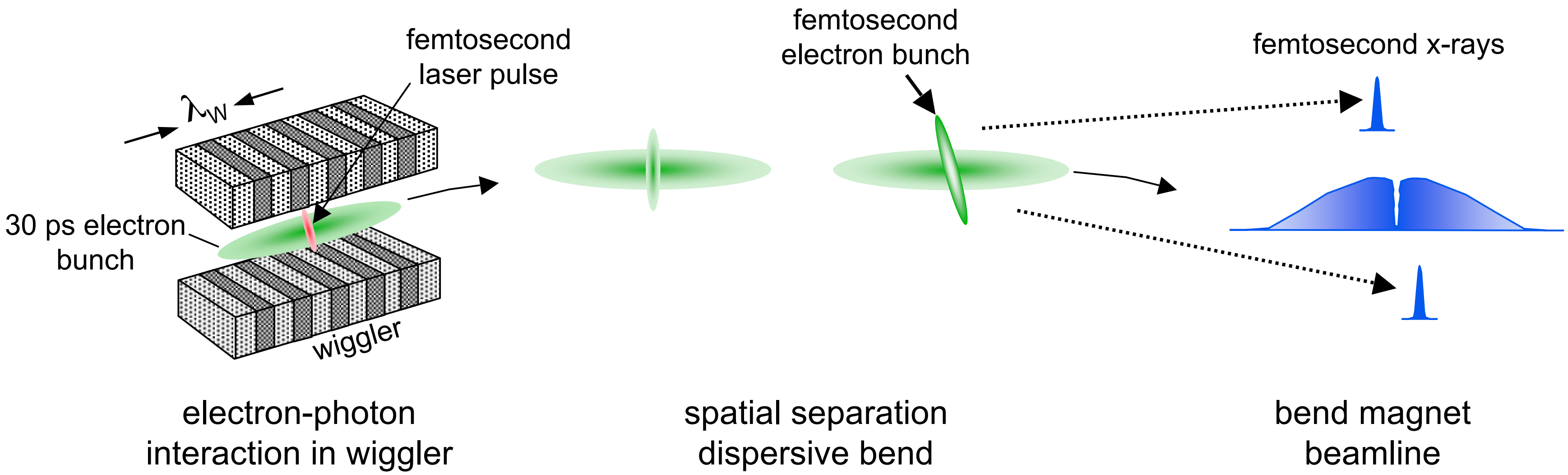


A. Lindenberg

- Implies molecular re-orientation around injected charge with similarities to thermally induced changes

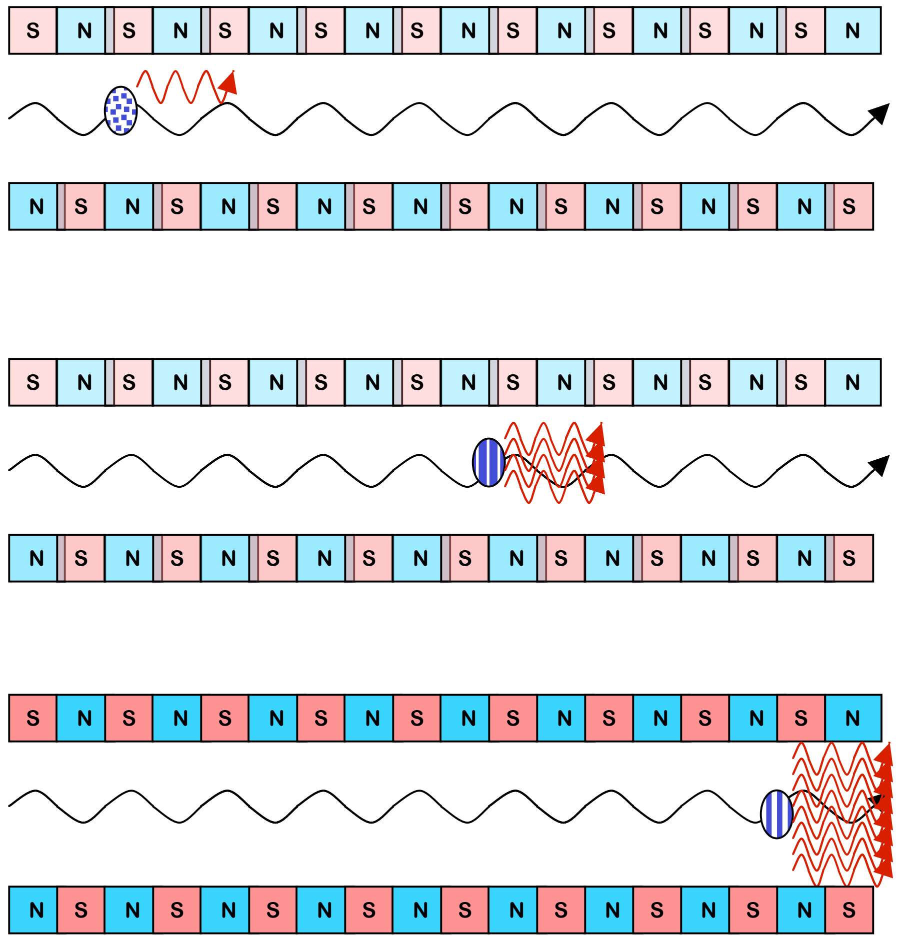
Laser-sliced x-ray pulses from synchrotrons are used as tunable probes of HED matter



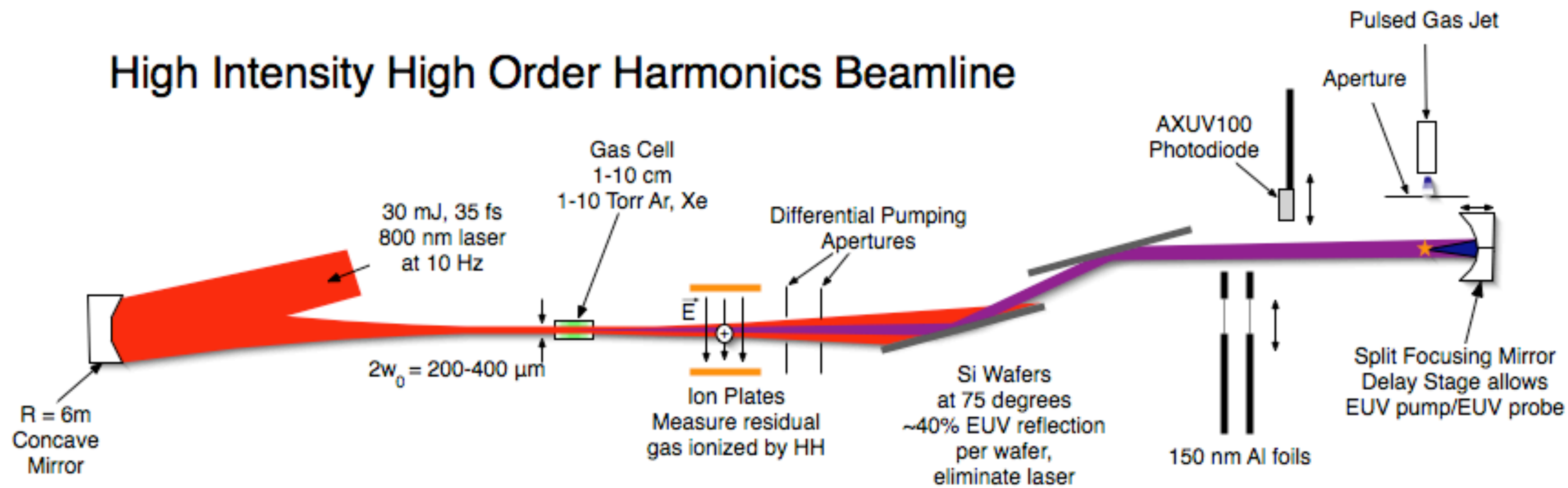


Zholents and Zolotorev, Phys. Rev. Lett., 76, 916,1996

X-Ray FELs produce x-ray pulses: eventually may be tunable for spectroscopy



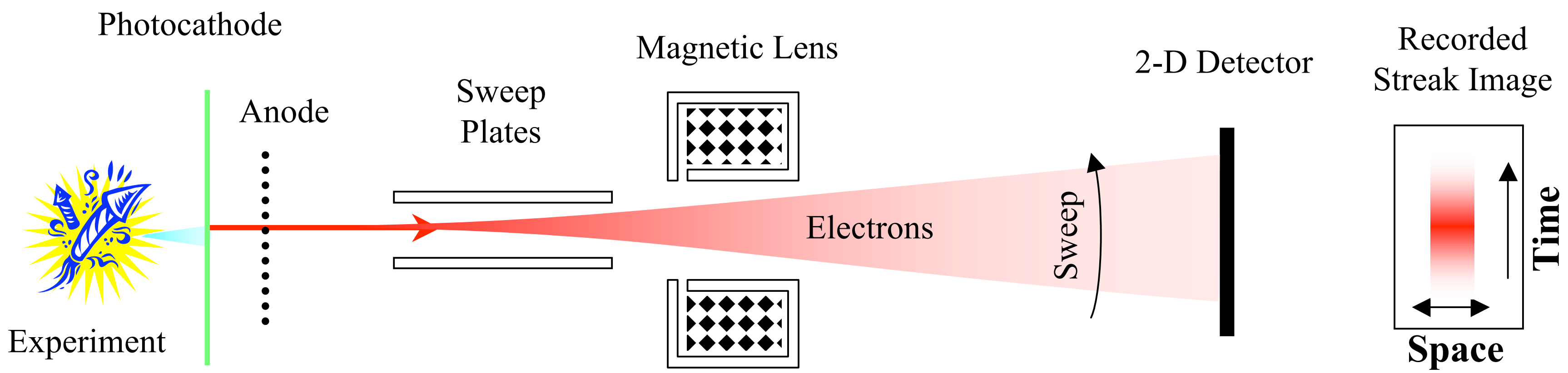
High-order harmonic radiation from multi-TW lasers produces intense soft x-ray fluxes for pump-probe HED science



- tunable soft x-ray peak power > 1 MW
- beam divergence < 1 mrad
- shot to shot fluctuations < 10%
- pulse length < 30 fs
- spatial and temporal coherence
- examine non-linear phenomena

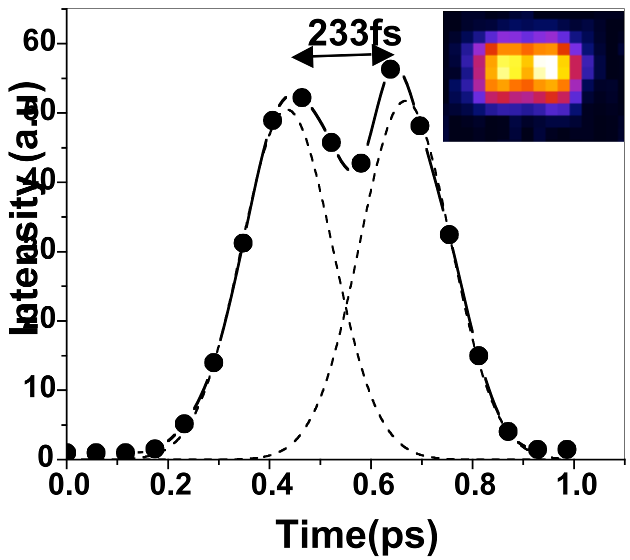
Allison, Belkacem,
Hertlein, VanTilborg

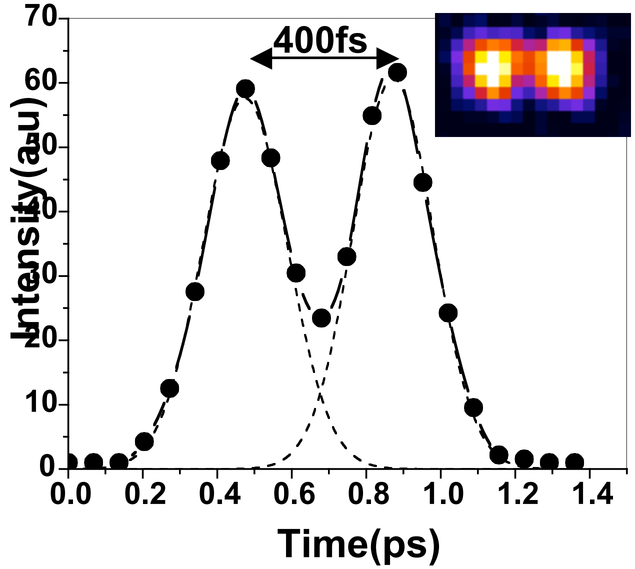
Ultrafast "x-ray streak cameras" enable high-speed recording of atomic dynamics



With space (1d) and time resolution, can record changing spectral response

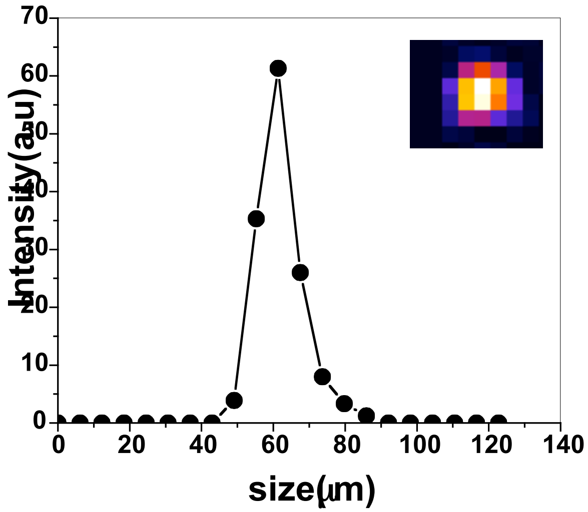
Fastest streak cameras can resolve << picosecond





Dynamic mode

1000 shots



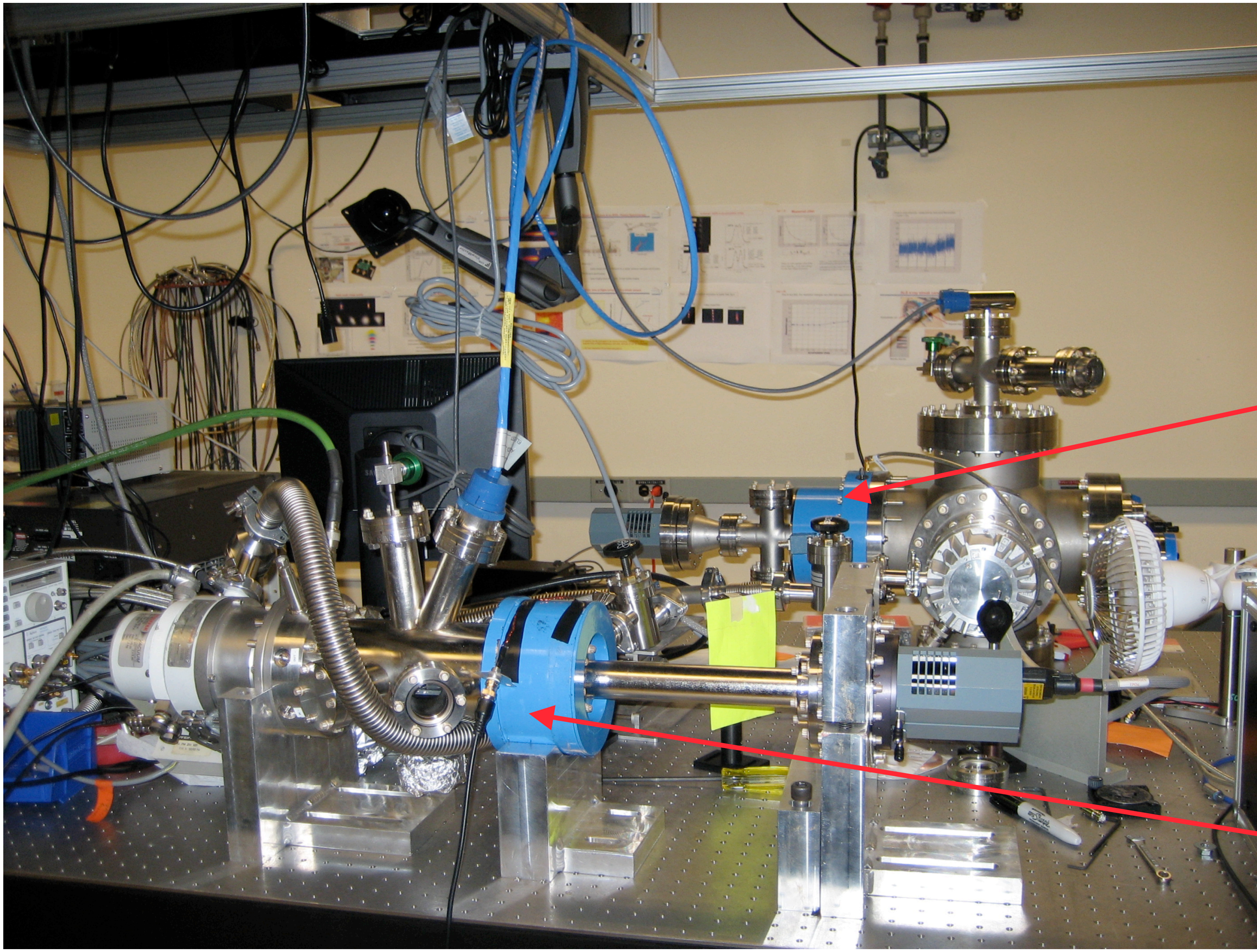
Static mode

1000 shots

Au photocathode

Jun Feng, Howard Padmore LBNL

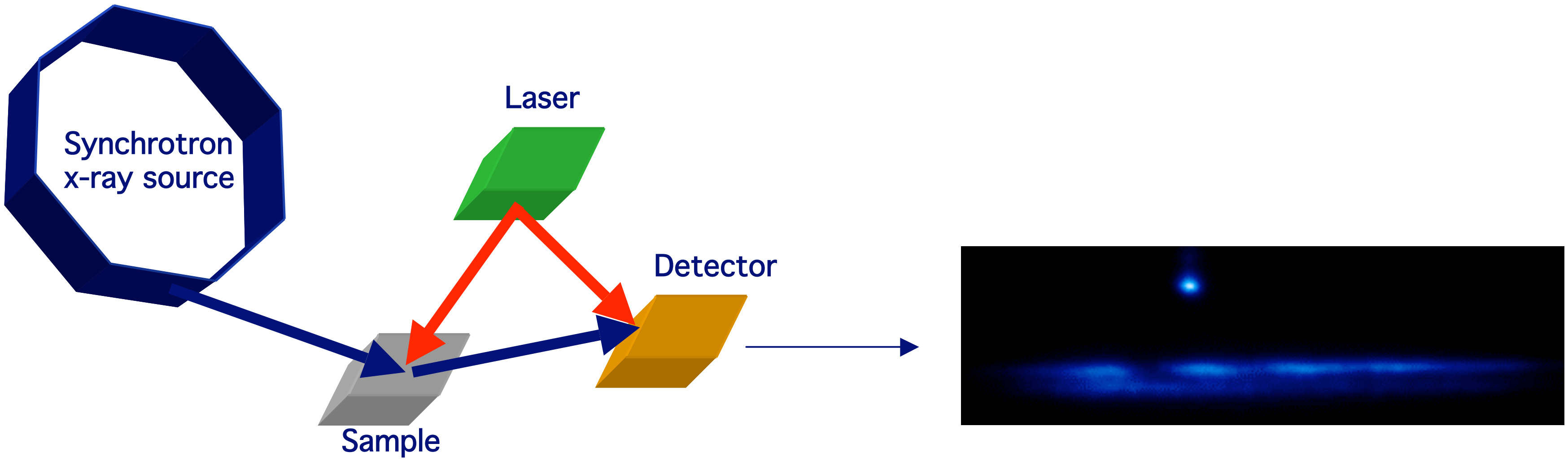
Ultra-fast X-ray Streak Cameras at the ALS

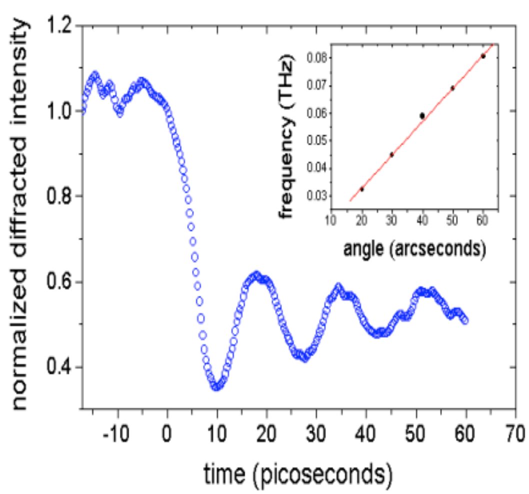


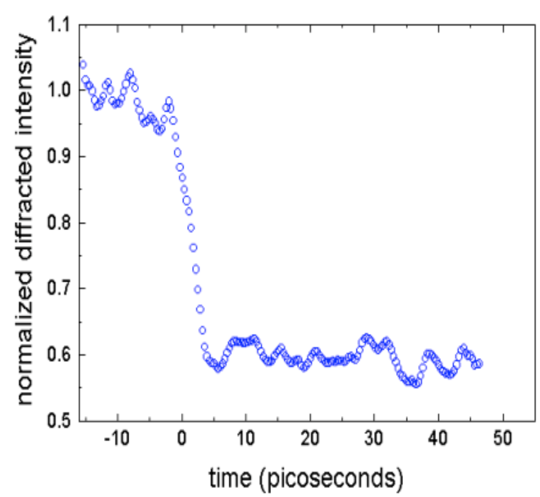
Transmission Streak Camera

Reflection Streak Camera

Laser-generated strain, bond-breaking, and hot electron-phonon coupling can initiate a solid-to-liquid phase transition which can be probed by ultrafast x-ray scattering

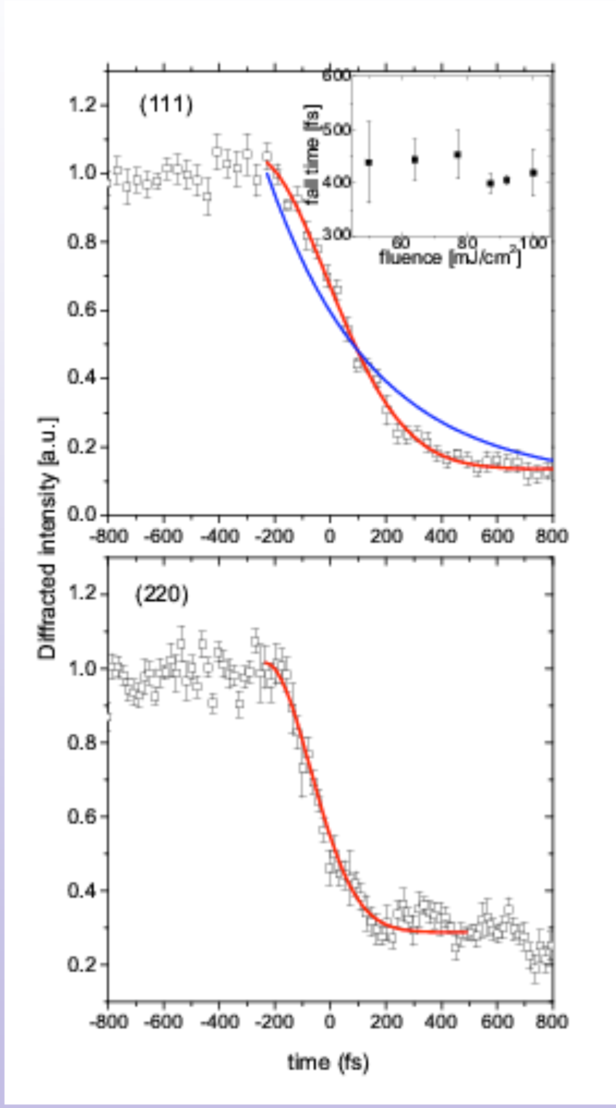






Lindenberg et al., Phys Rev. Lett. 84, 111 (2000)

Disordering of a lattice through bond-breaking observed at even shorter times at the SPPS



- (111) and (220) reflections measured
- non-thermal melting observed

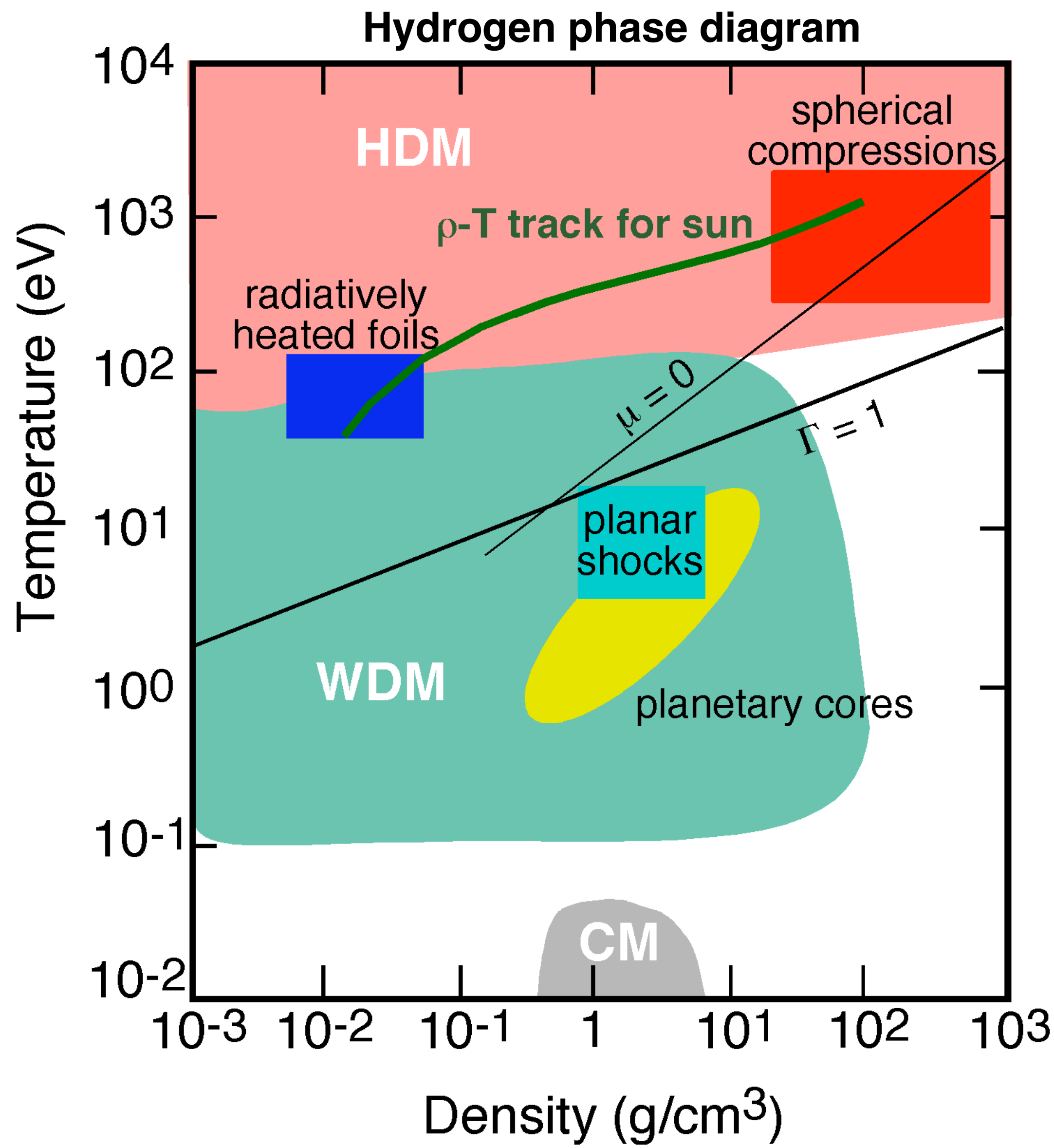
$$\frac{\tau_{(111)}}{\tau_{(220)}} = 1.6 \pm 0.2 = \frac{G_{(220)}}{G_{(111)}}$$

$$\sqrt{2^2+2^2+0^2}/\sqrt{1^2+1^2+1^2} = \sqrt{\frac{8}{3}}$$

SPPS Collaboration

High Energy Density Matter occurs widely

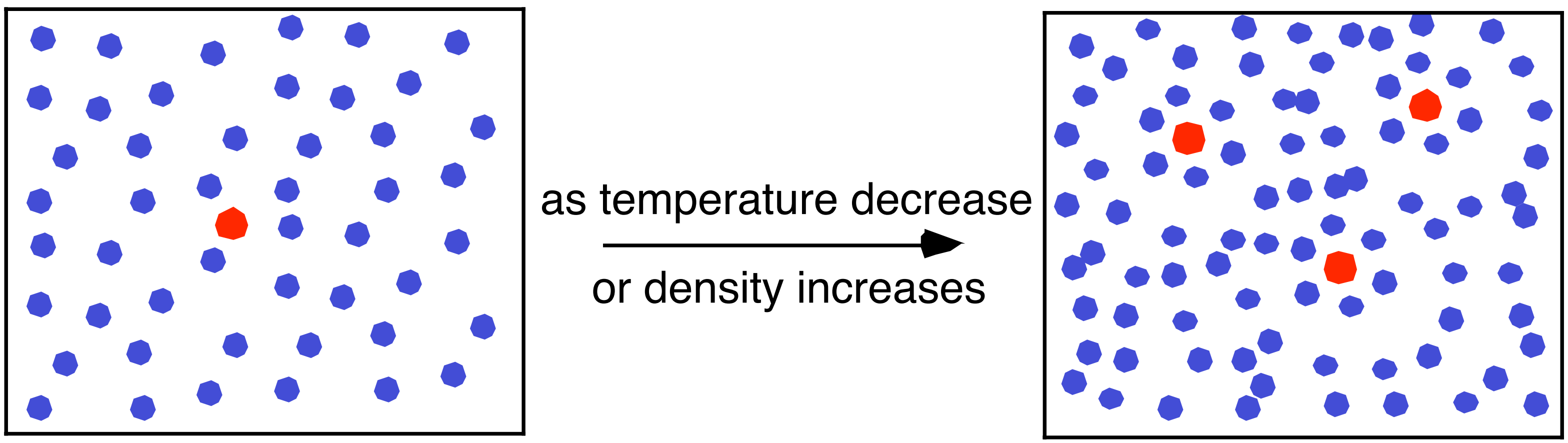
- **Hot Dense Matter (HDM) occurs in:**
  - Supernova, stellar interiors, accretion disks
  - Plasma devices: laser produced plasmas, Z-pinches
  - Directly and indirectly driven inertial fusion experiments
- **Warm Dense Matter (WDM) occurs in:**
  - Cores of large planets
  - X-ray driven inertial fusion experiments



The defining concept of warm dense matter (WDM) is coupling

weakly coupled plasmas

- plasma seen as separate point charges
- plasma is a bath in which all particles are treated as points

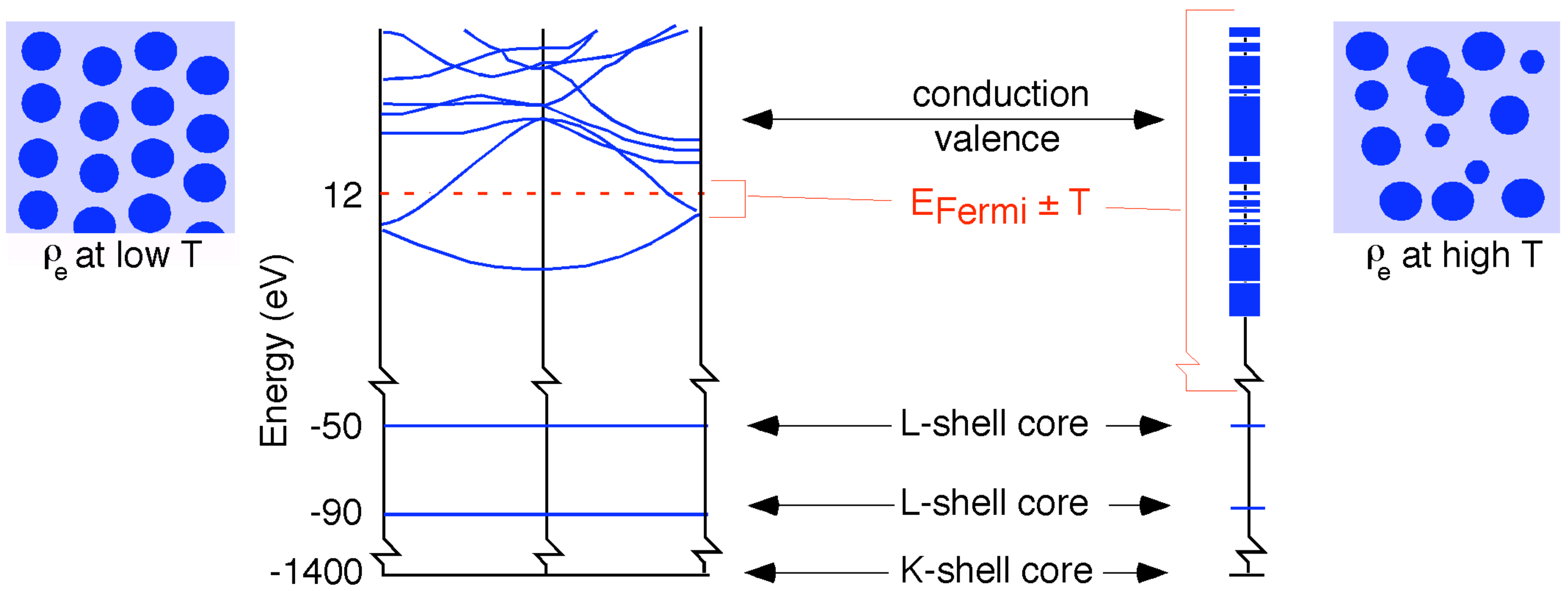


when either ρ increases or T decreases, $\Gamma > 1$

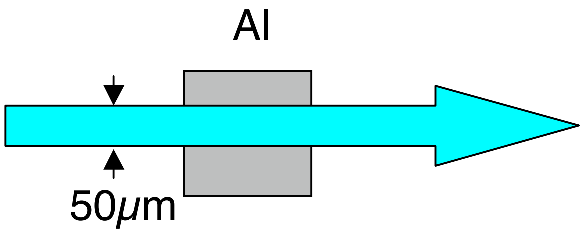
- particle correlations become important
- energy levels shift and ionization potentials are depressed

WDM is defined by temperature relative to the Fermi energy

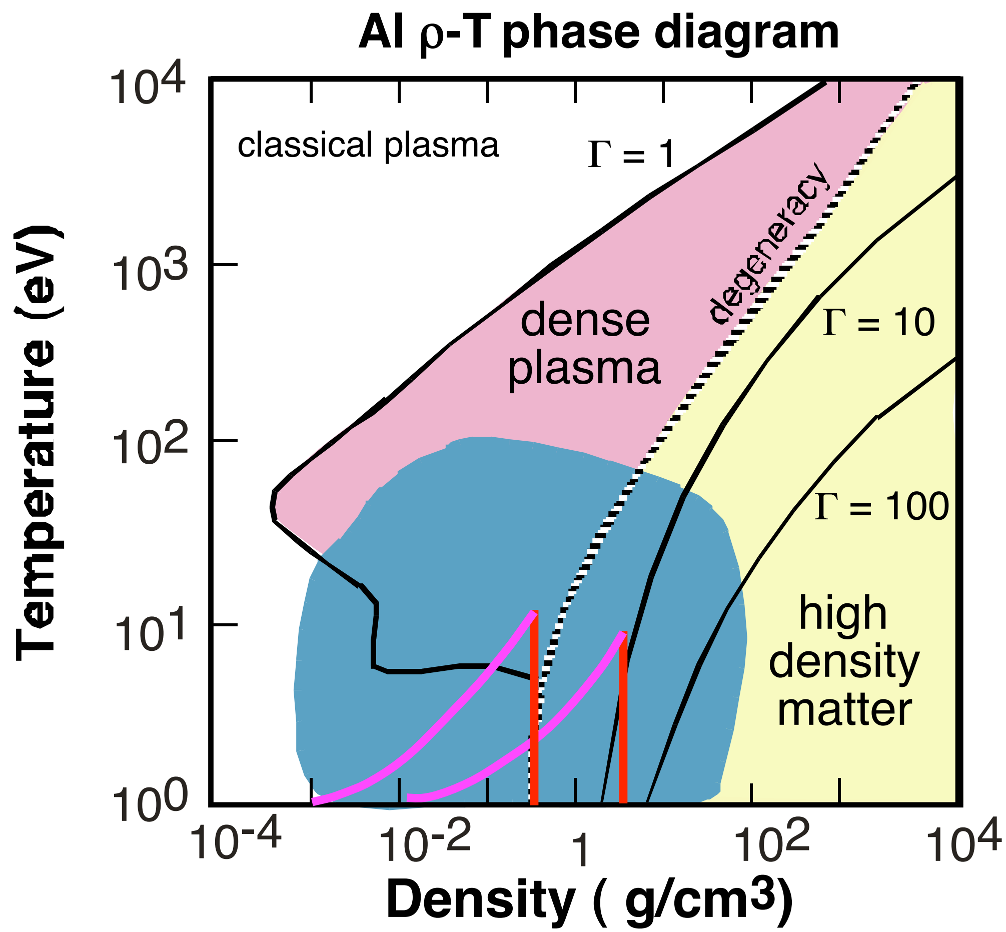
- Fermi energy, E_{Fermi}, = maximum energy level of e^- in cold matter
- When $T \ll E_{Fermi} = T_{Fermi}$ standard condensed matter methods work
- When $T \sim T_{Fermi}$ one gets excitation of the core
  - Ion - e^- correlations change and ion-ion correlations give short and long range order



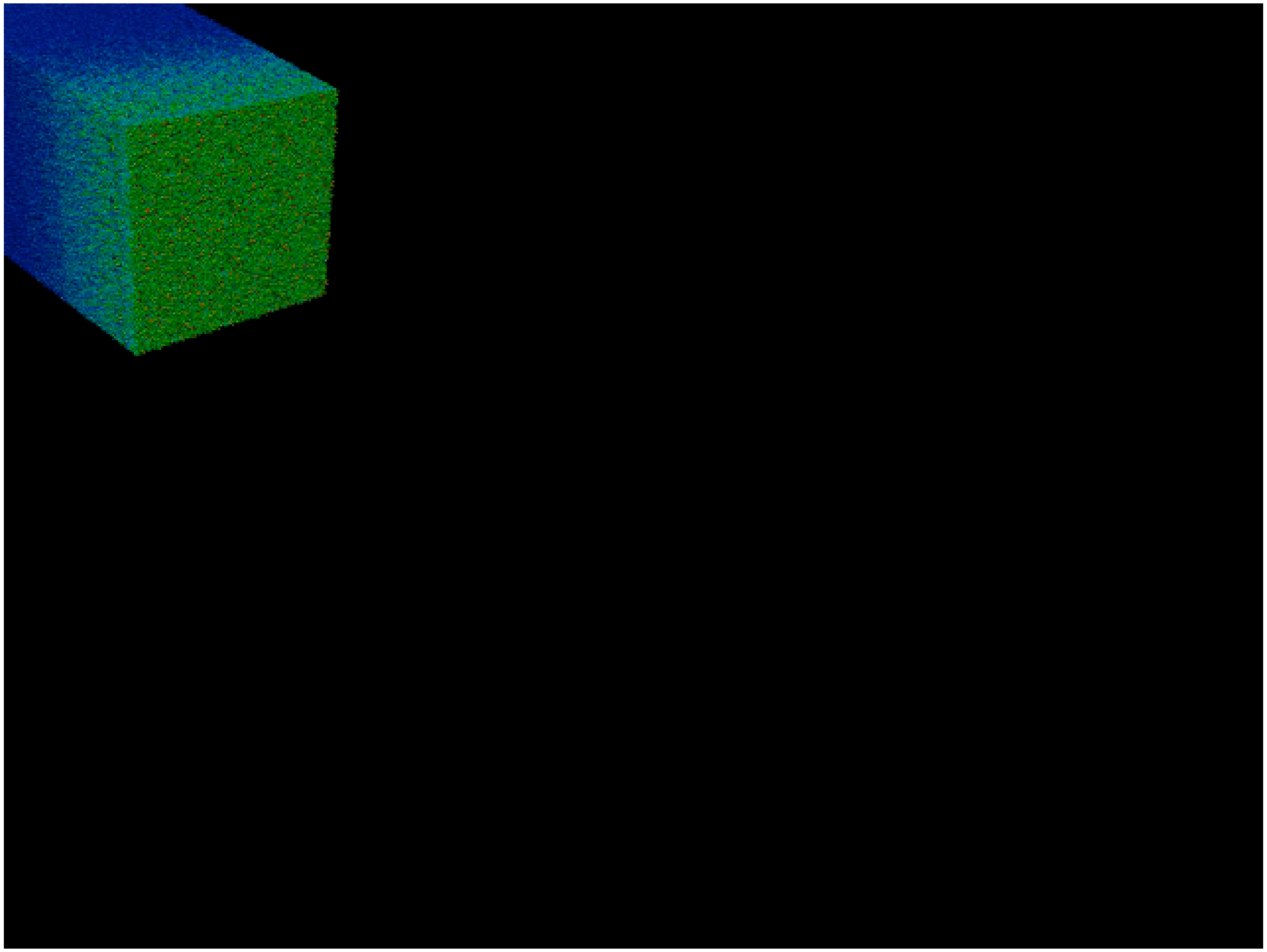
WDM, created by isochoric heating using short pulses, will isentropically expand sampling phase space



- XFEL can heat matter rapidly and uniformly to create:
  - Isochores (constant ρ)
  - Isentropes (constant entropy)
- Using underdense foams allows more complete sampling
  - Isochores (constant ρ)
  - Isentropes (constant entropy)

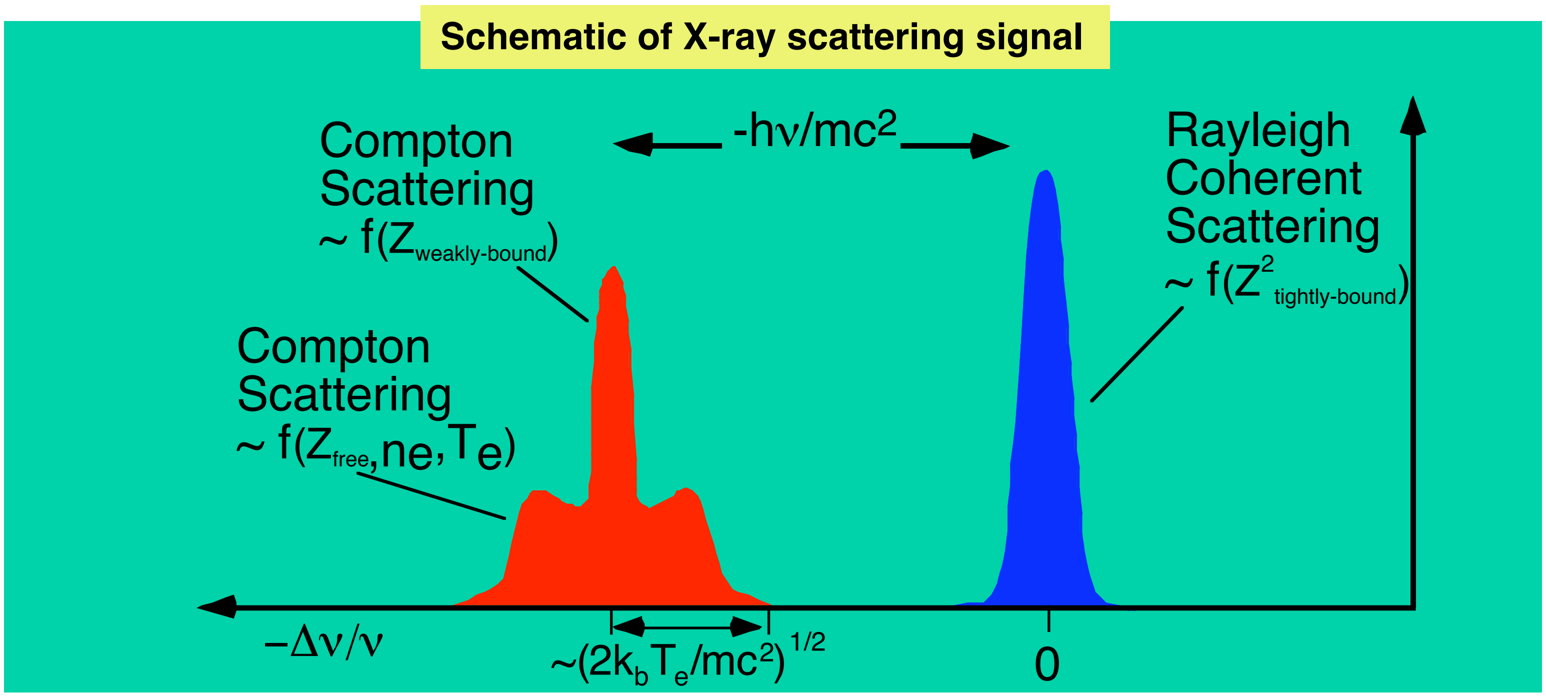


Ablation of a surface under high energy flux



G. Gilmer, B. Sadigh, LLNL

Thomson scattering enables direct determination of both material *and* plasma properties

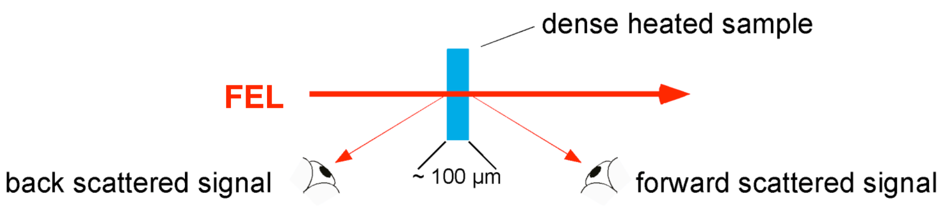


- 25 eV, 4×10^{23} cm^{-3} plasma XFEL produces10^4 photons from the free electron scattering

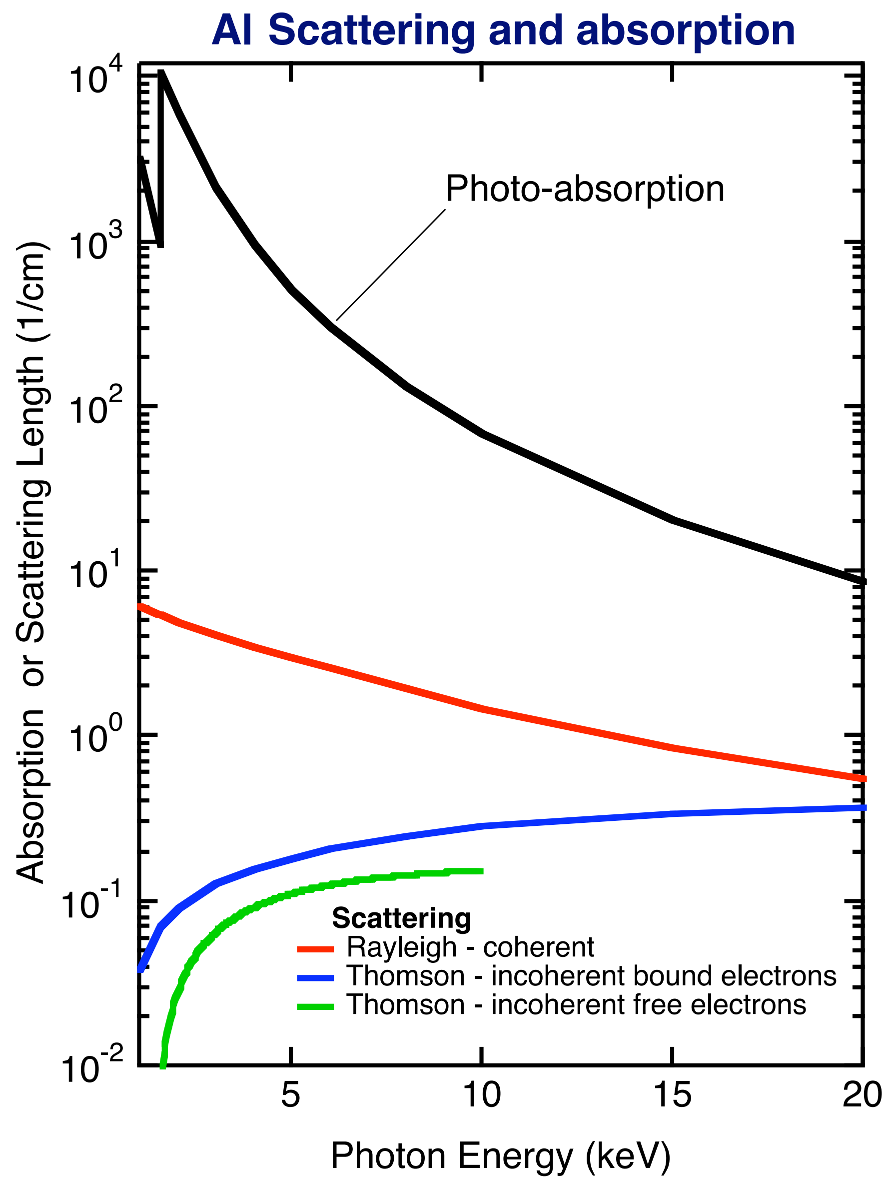
- Can obtain temperatures, densities, mean ionization, velocity distribution from the scattering signal

By varying the scattering angle, collective modes of dense matter are probed

X-rays provide a unique probe of HED matter



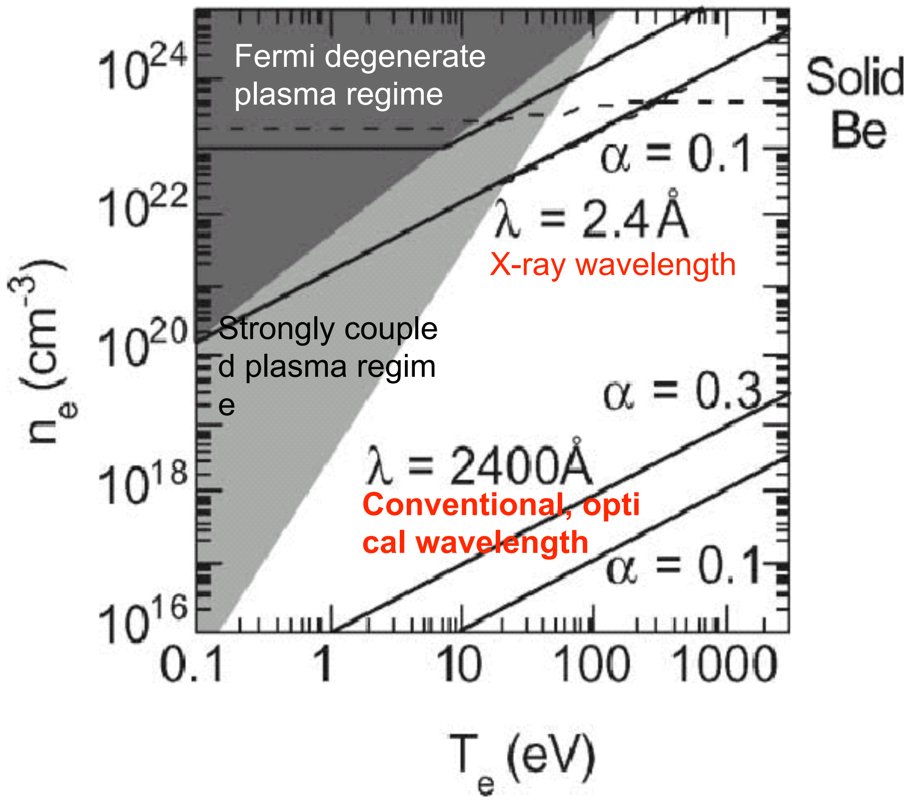
- Due to absorption, refraction, & reflection, visible lasers cannot probe high density
- X-ray scattering from free electrons provides a measure of the T_e, n_e, $f(v)$, and plasma damping
- x-ray FEL scattering signals will be well above noise for HED matter

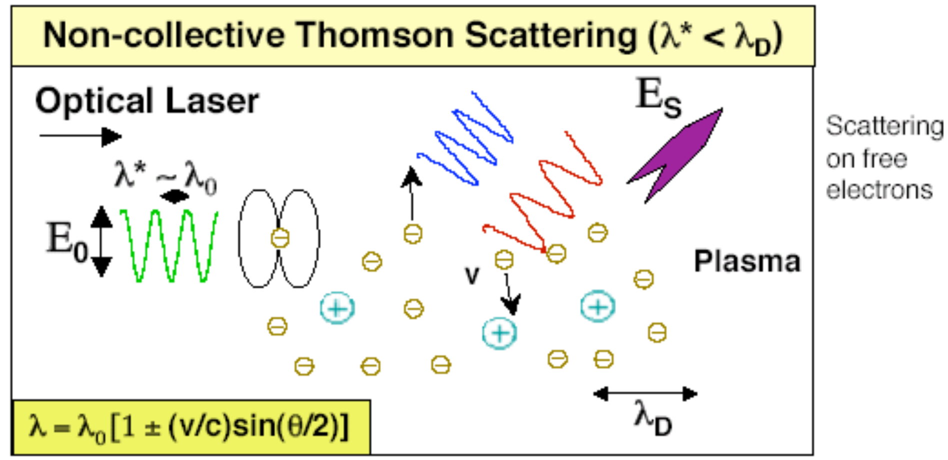


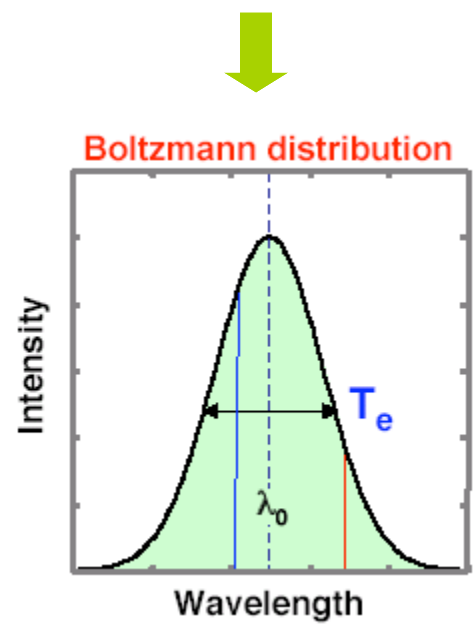
Compton/Thomson scattering with optical probe

O. L. Landen et al. JQSRT (2001)

θ=180°, α=0.3







Fermi degenerate plasma regime: $T_e < T_F$

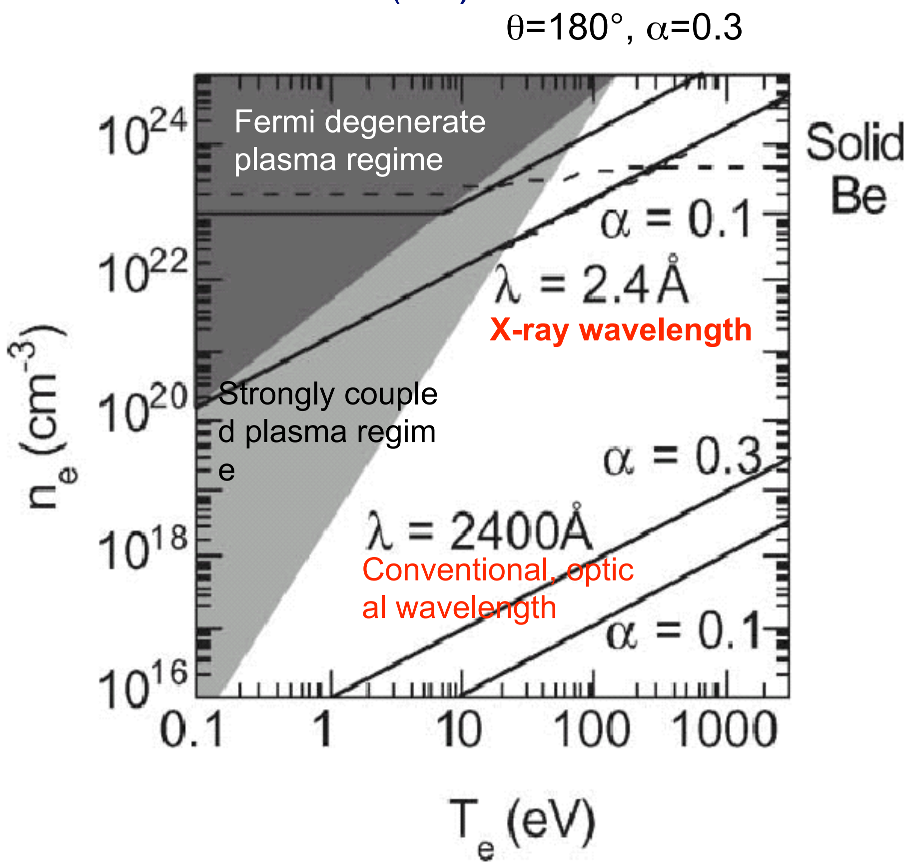
Strongly coupled plasma regime : $T_e > T_F$, $\Gamma_{ee} > 1$

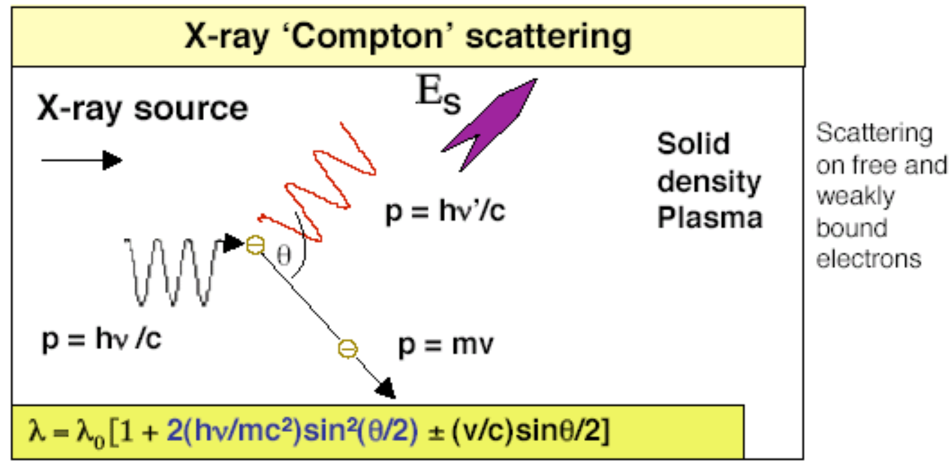
Γ_{ee}=Coulomb potential energy/Kinetic energy of free electrons

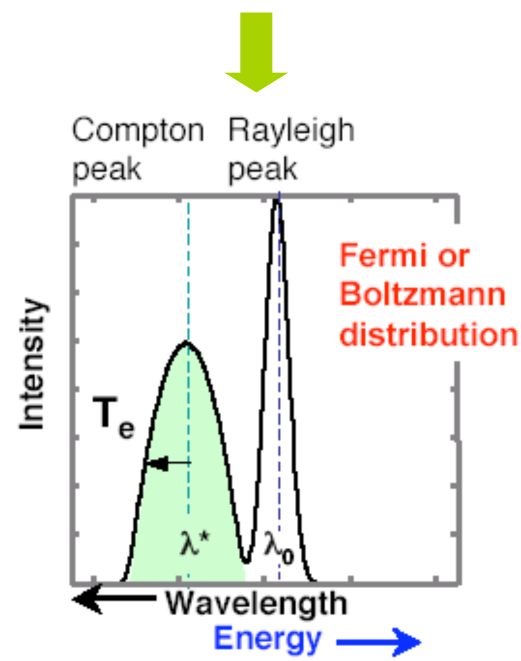
Ideal plasma: $\Gamma_{ee} < 1$

Compton/Thomson scattering with x-ray probe in dense matter

O. L. Landen et al. JQSRT (2001)







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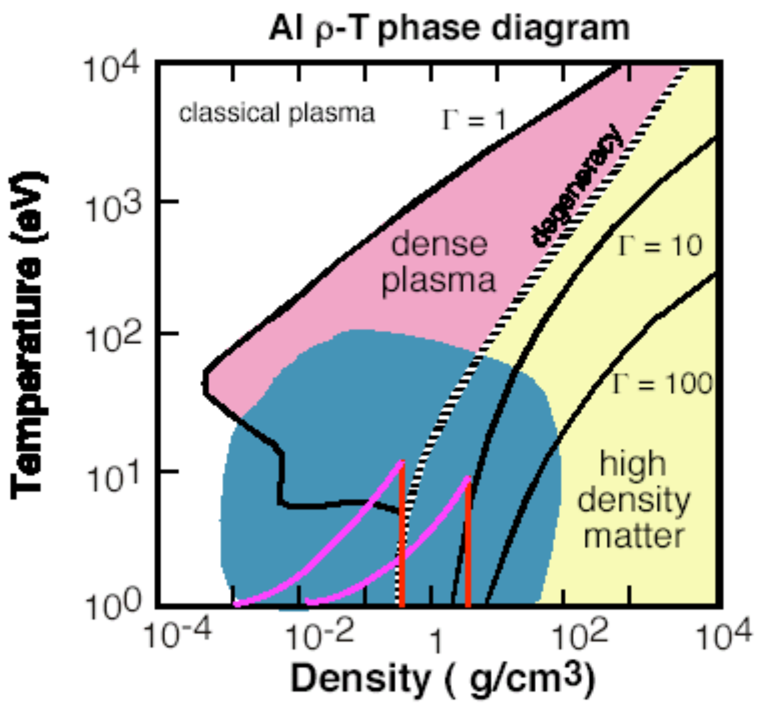
Scattering regimes in the ρ-T plane



In dense plasmas

- standard theoretical approaches fail

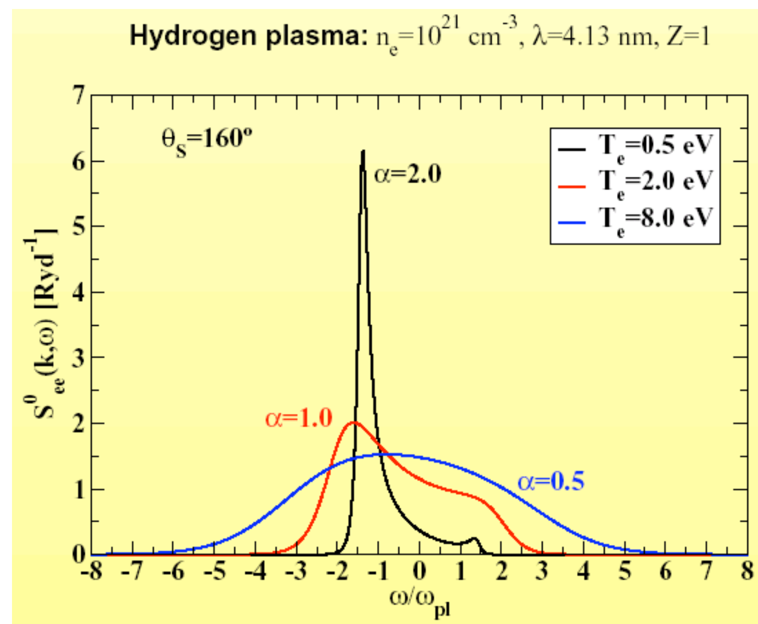
- theoretical uncertainties are large

Collective scattering in dense plasmas

- probes transition region

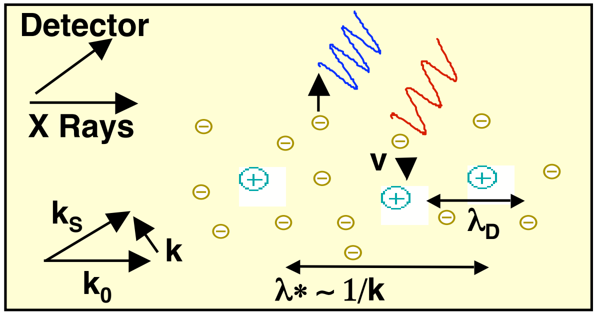
X-ray source

- penetrates dense plasmas



Forward scattering and plasmon in dense matter

Forward scatter on Plasmons



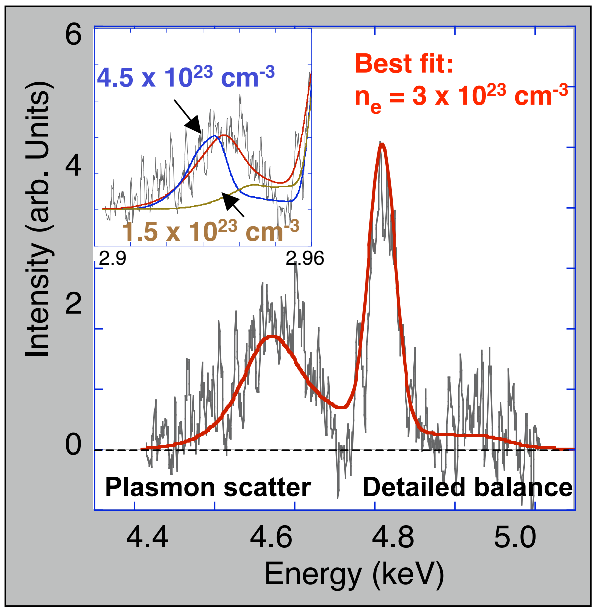
Scattering parameter α

$\alpha = 1/(k\lambda_D) \sim \lambda_L / (4\pi\lambda_D \sin(\theta/2)) \rightarrow \lambda^*/\lambda_D$

$\alpha > 1$: Collective regime, $\lambda^* > \lambda_D$,

Orderly oscillatory behavior under the long-range Coulomb f orces.

<u>**The density fluctuation in the plasma behave collectively and oscillate around ω_p.**</u>



With x-ray probe for WDM, strong asymmetry or almost gone of blue-shifted peak.

<u>**From the plasmon peak, we can have better accurate info rmation about T_e!!**</u>

From the fluctuation-dissipation theorem

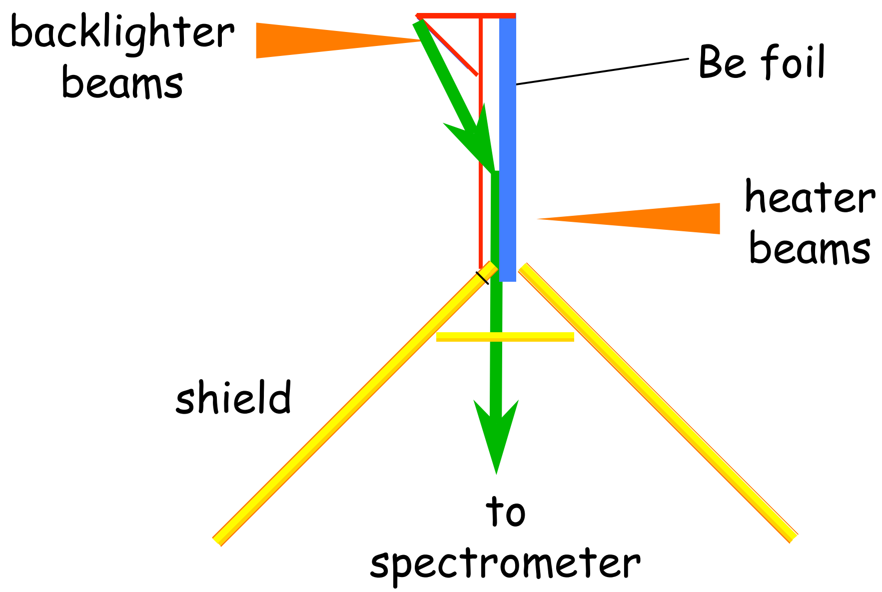
$$S(k,\omega) = \frac{1}{2\pi N}\int e^{i\omega t} < \rho_e(\vec{k},t)\rho_e(-\vec{k},0) > dt,$$

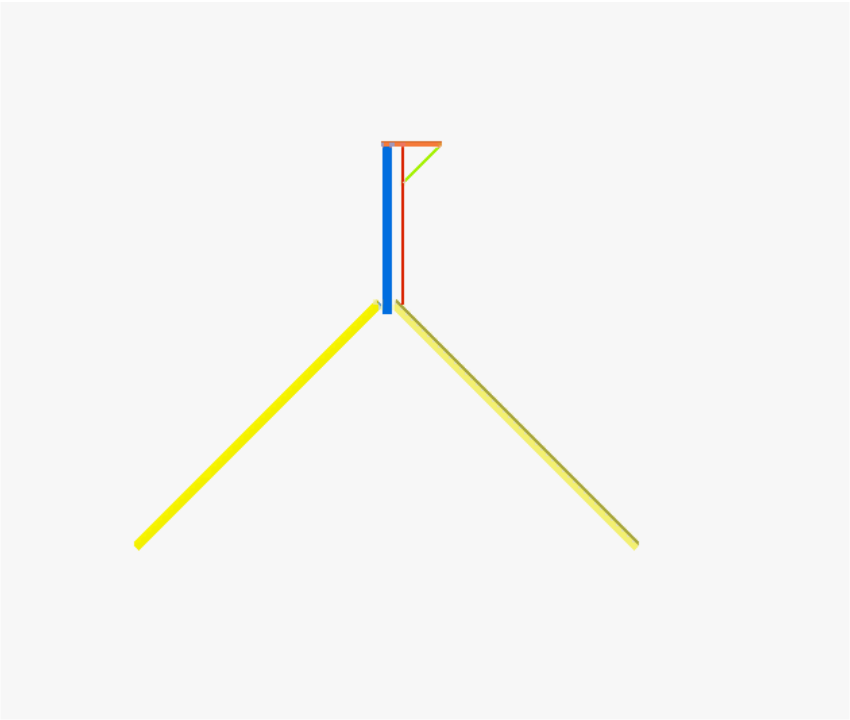
$$S(k,\omega) = -\frac{\varepsilon_0 \hbar k^2}{\pi e^2 n_e}\frac{1}{1-e^{\hbar\omega/k_B T}}\operatorname{Im}\varepsilon^{-1}(k,\omega),$$

$$\Rightarrow \quad \frac{S(k,\omega)}{S(-k,-\omega)} = \exp\left(-\frac{\hbar\omega}{k_B T}\right) \quad \Rightarrow$$

<u>Sensitive to T_e</u>

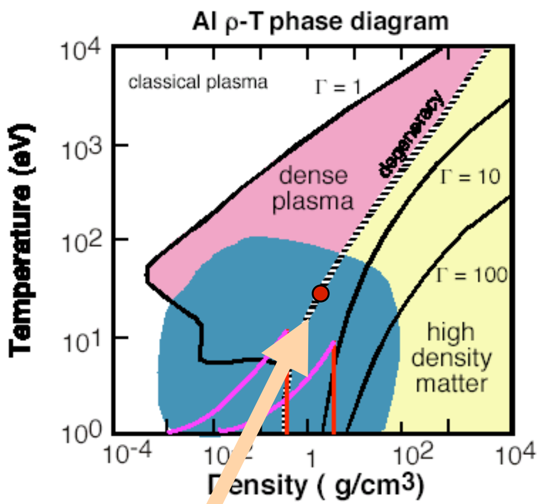
Current Thomson scattering experiments are done at large laser facilities





Haeja Lee UCB

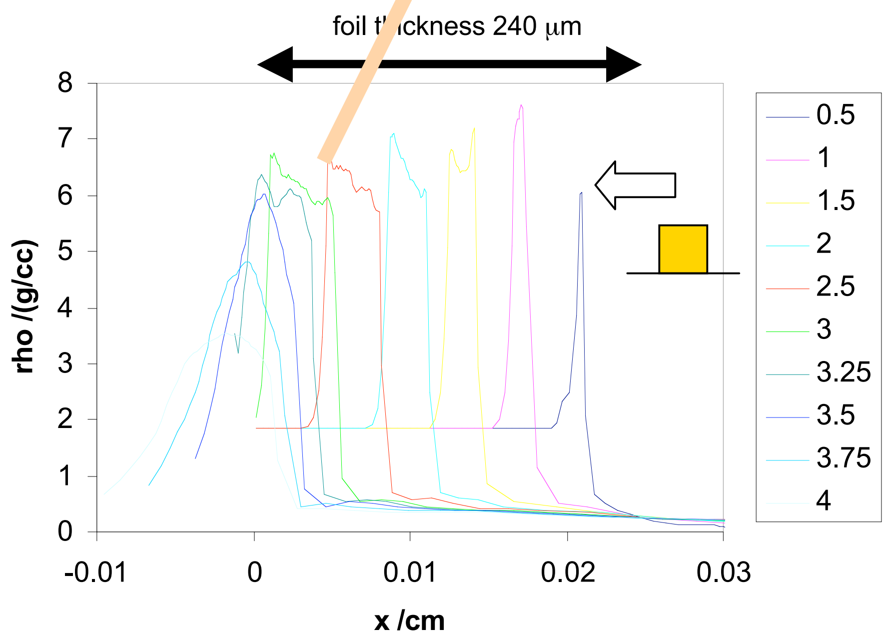
1-D HYADES Code predicts plasma conditions under shock propagation

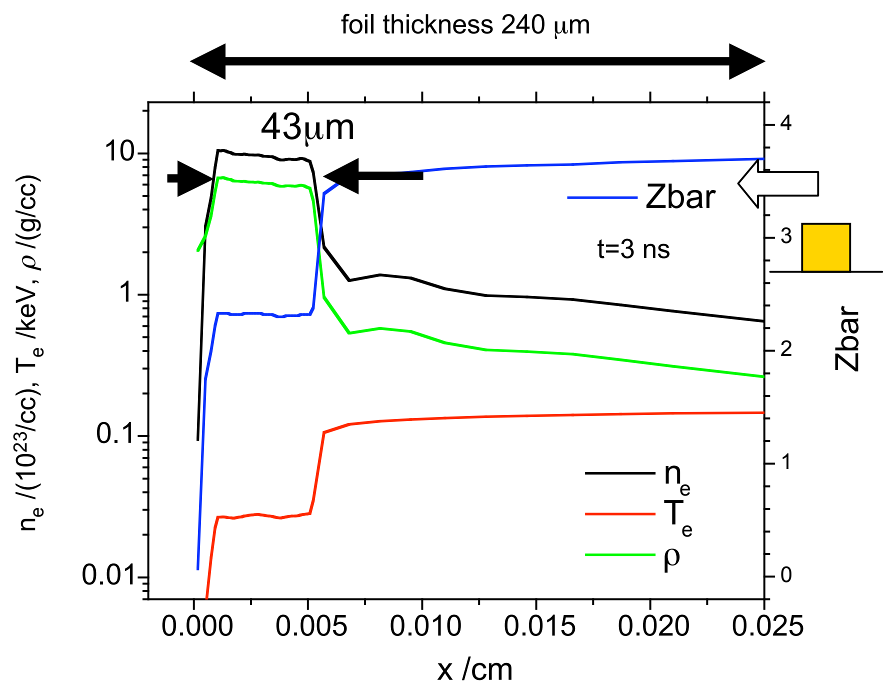


Omega laser = 17 beams, 480 J each, total energy ~8.7 kJ,

Target: Be-foil, thickness 0.24 mm
Laser intensity: 3×10^{14} W/cm^2
Pulse duration: 3 ns

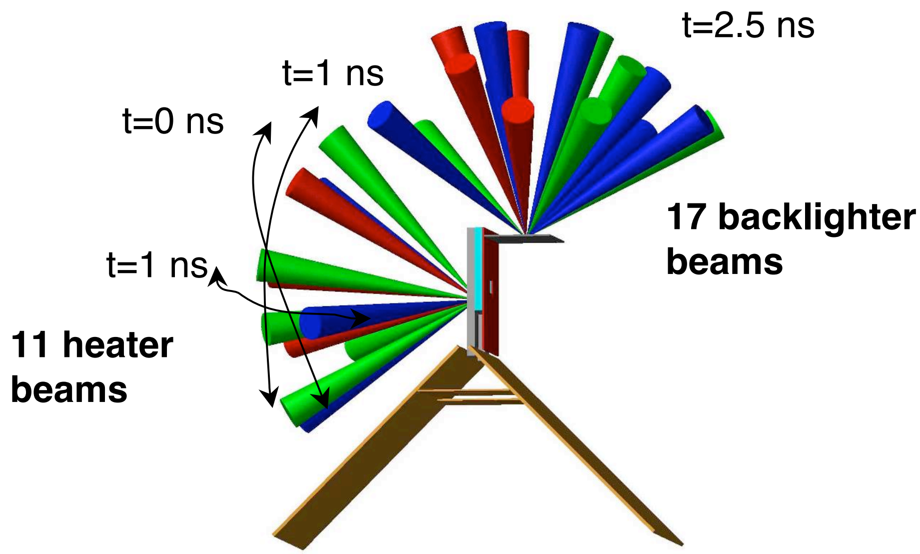
WDM with ~43 μm depth is generated over 500 ps with uniform condition.

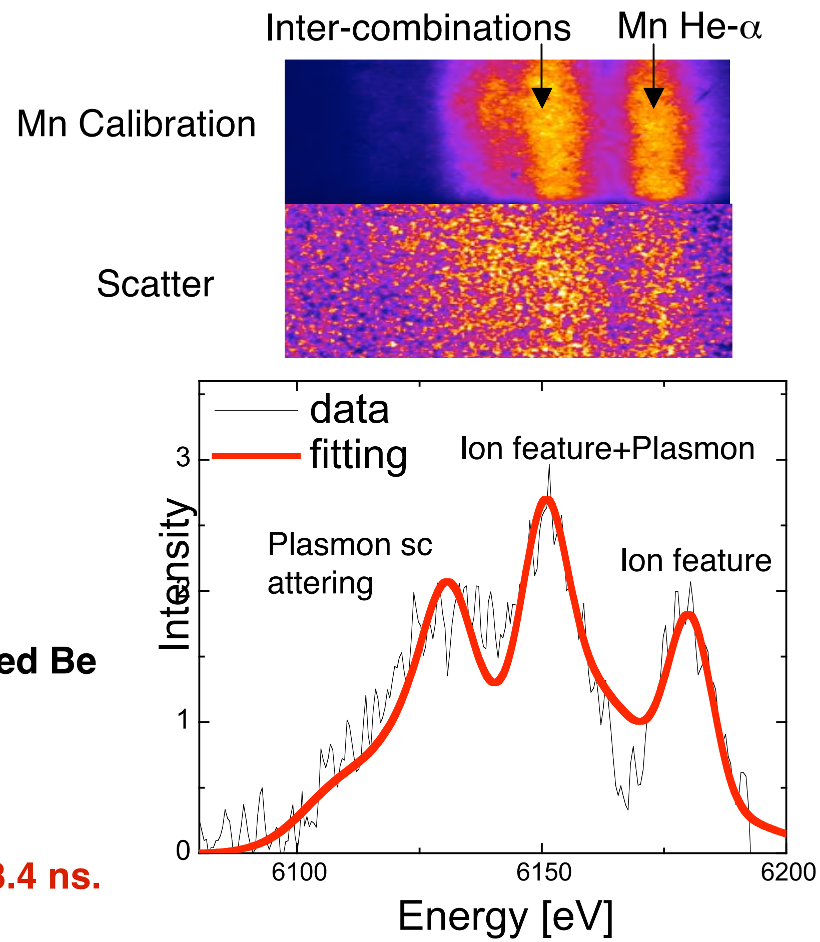




X-ray Thomson scattering on compressed Be

NLUF experiments in May 2007 measured x-ray scattering on compressed Be





We obtained plasmon scattering from shock compressed Be

- **position of the plasmon resonance yields density**

~ n_e=1×10^{23} cm^{-3}, T_e=10 eV at 3 ns

2 ns drive beams at t = 0; analyze plasma between 2.6-3.4 ns.

Thomson scattering at large laser facilities or XFELs ?

- for x-ray pulse backlighting of warm matter on high-energy laser systems, we use multiple laser beams with about 10,000 J in a few ns, for pumping a plasma on a surface that radiates K-α and He-α x-rays

- this converts to about 1 J of x-ray photons radiated into 4π

- there is then about 1 mJ for use in illuminating the sample, within the collected solid angle

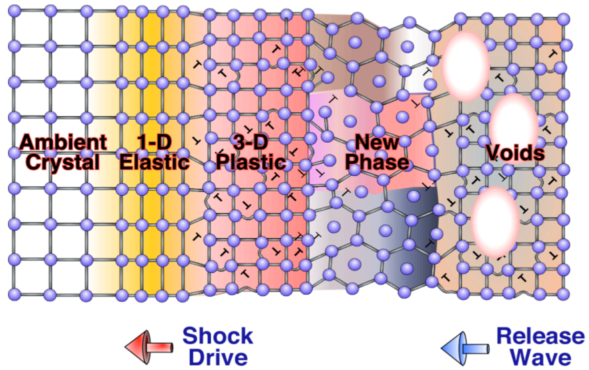
- this probe x-ray beam compares well in energy per pulse with the LCLS per pulse energy, which has 1 mJ

- LCLS pulses will be more collimated, narrower BW, and shorter in duration (~ 200 fs)

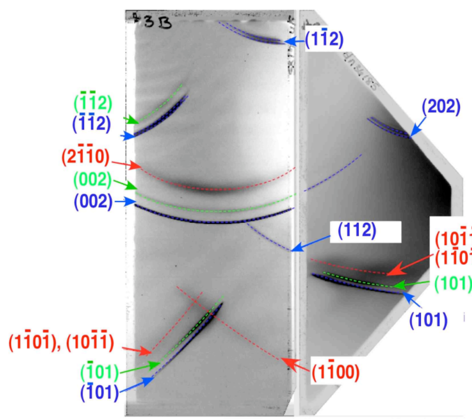
Materials science and lattice dynamics at ultrahigh pressures and strain rates define a frontier of condensed matter science

The National Ignition Facility

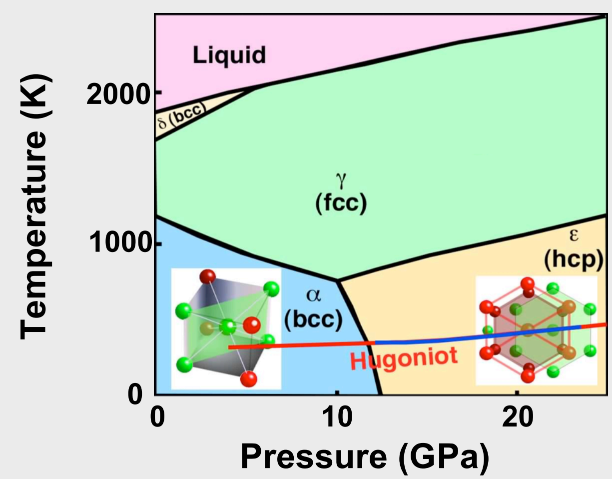
Lattice Dynamics



Dynamic Diffraction Lattice Msmt



Phase Diagram of Iron



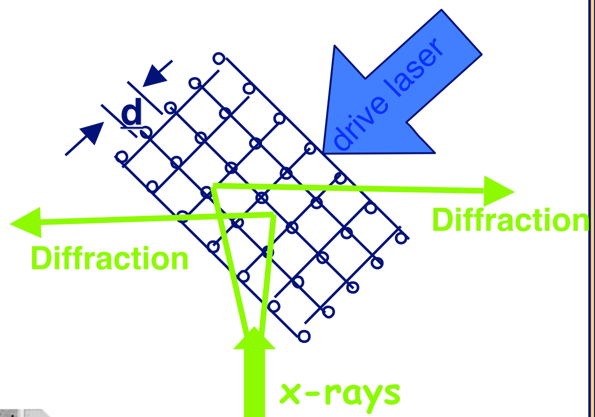
Unexplored regimes of solid-state dynamics at extremely high pressures and strain rates will be accessible on NIF

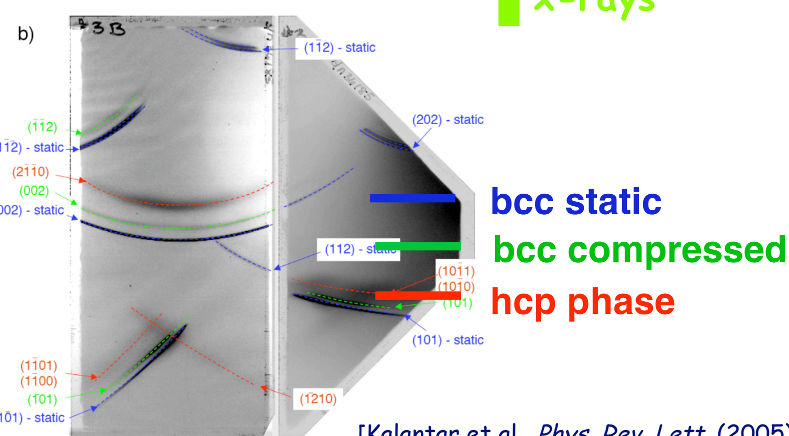
[D.H. Kalantar et al., PRL 95 ,075502 (2005); J. Hawreliak et al., PRB, in press (2006)]

Intense x-ray fluxes from LCLS will enable real-time *in situ* measurements of microstructure evolution at high pressure

What is the timescale of the bcc-hcp phase transformation in Fe?

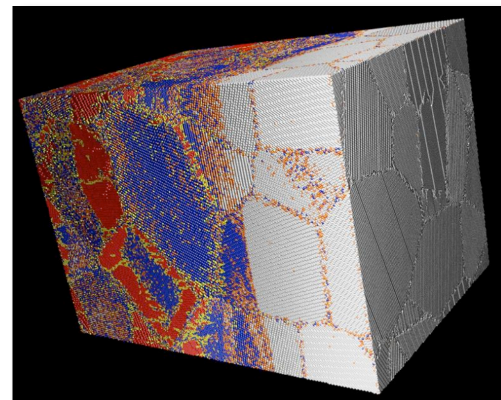
Current measurement limited to timescales >> psec



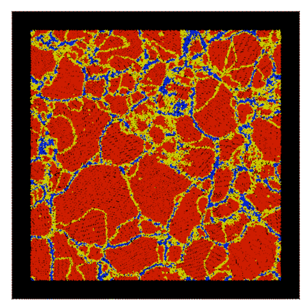


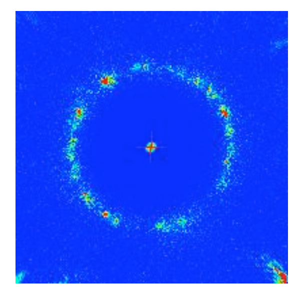
[Kalantar et al., *Phys. Rev. Lett.* (2005)]

Simulations predict subpicosecond phenomena observable using LCLS



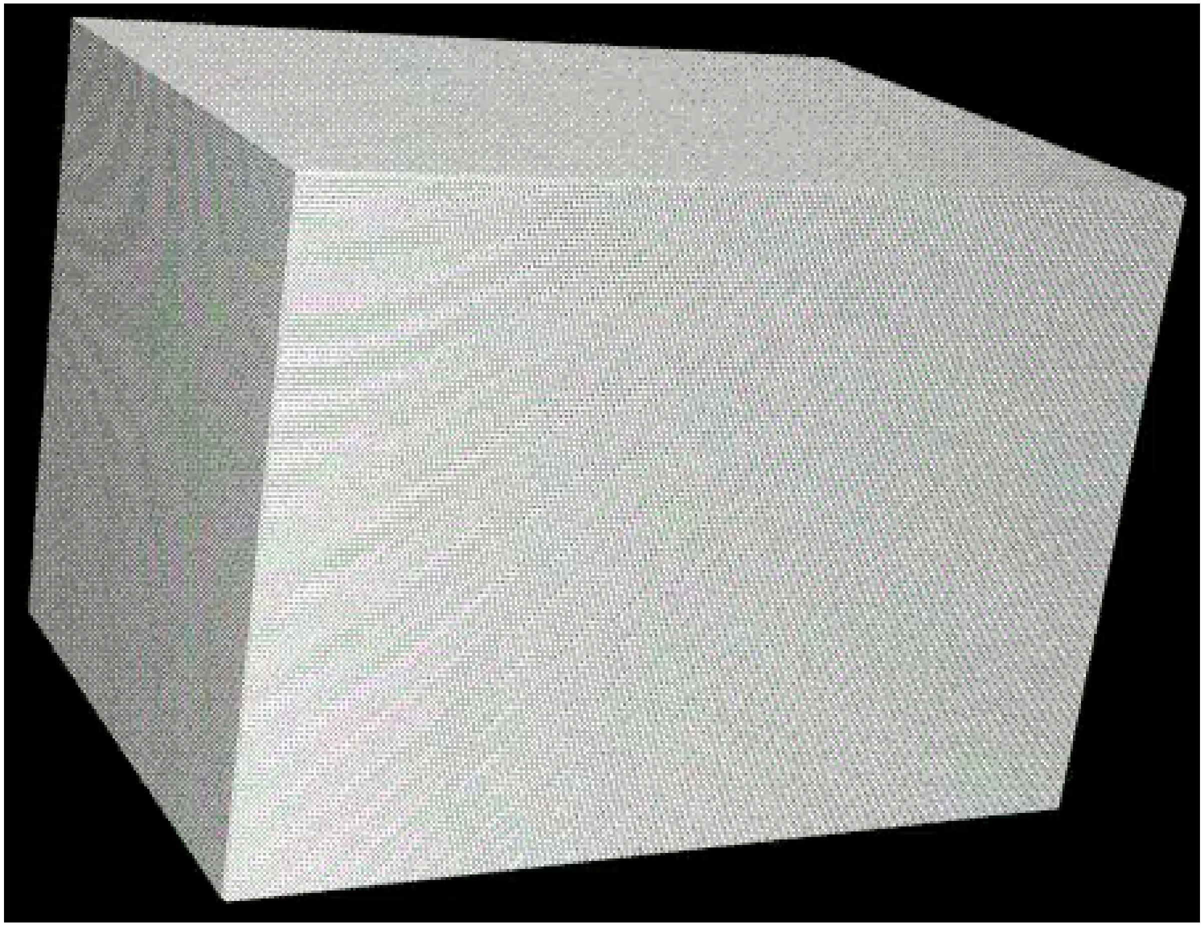
Kadau et al., *Science* (2002).



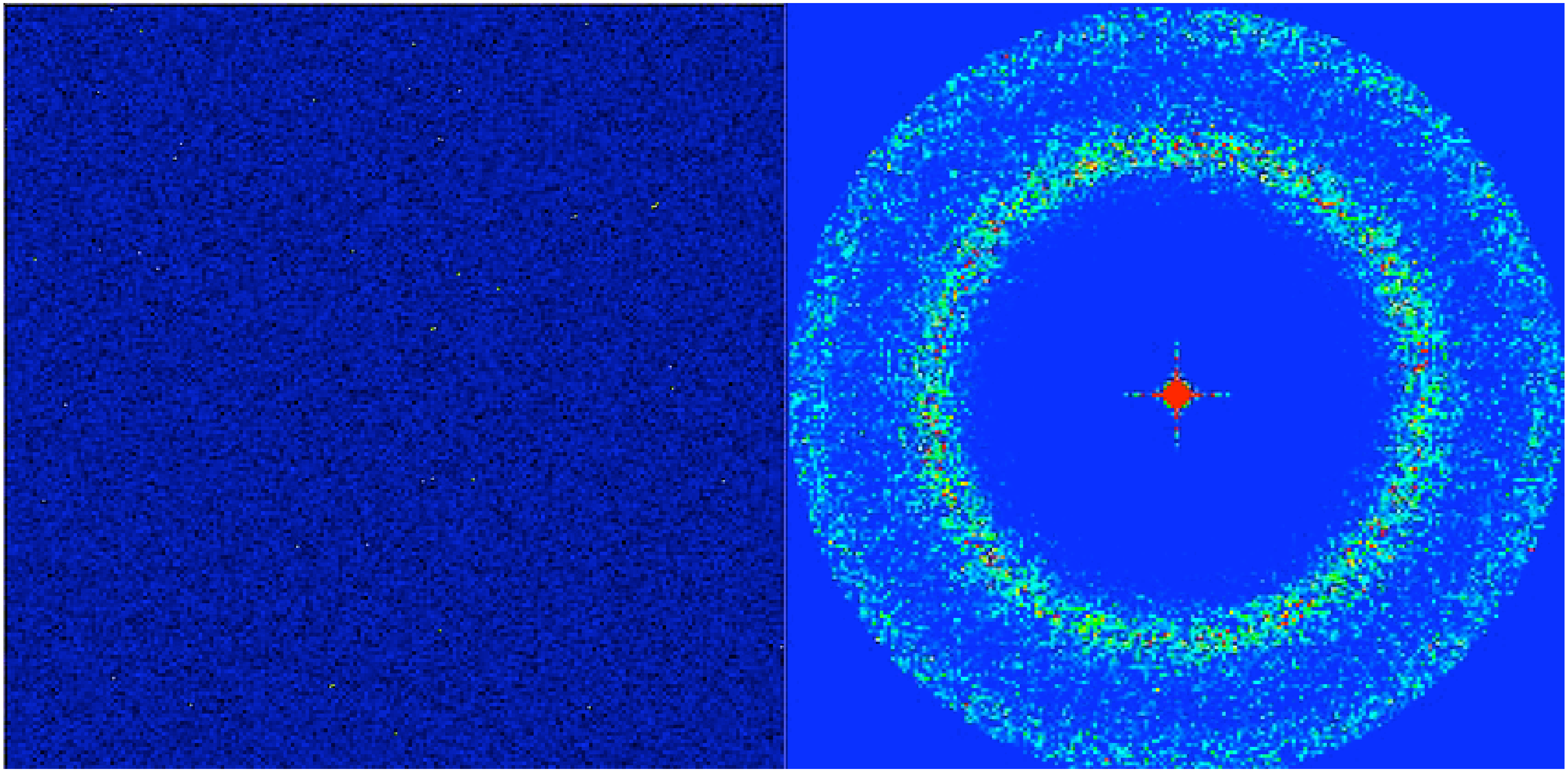


Molecular Dynamics simulations indicates shock-driven phase transitions take ~ 1 ps

Grey = static BCC   Blue = compressed BCC   Red = HCP

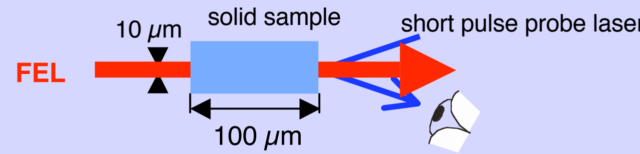


- 8 million atoms, total run time 10 ps (K. Kadau LANL)
- Require LCLS to time-resolve kinetics of the transition

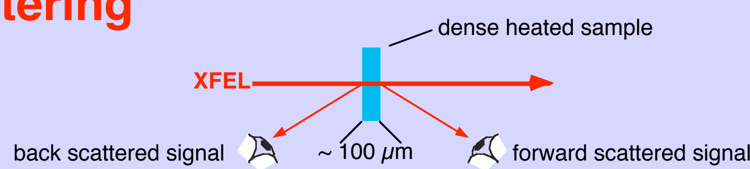


X-Ray FELs will enable a range of HED experiments
(talk by R.W. Lee)

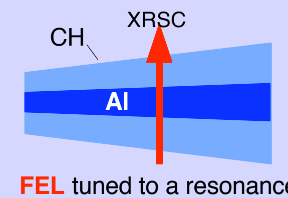
- **Creating Warm Dense Matter**
  - Generate ~ 10 eV solid density matter
  - Measure the equation of state



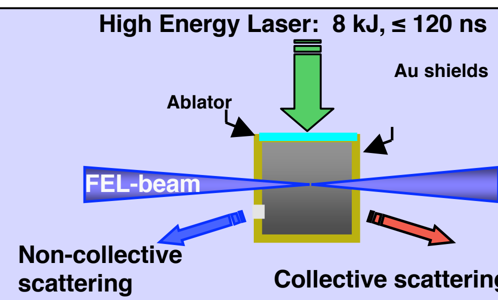
- **Probing dense matter with Thomson Scattering**
  - Perform scattering from solid density plasmas
  - Measure n_e, T_e, <Z>, f(v)



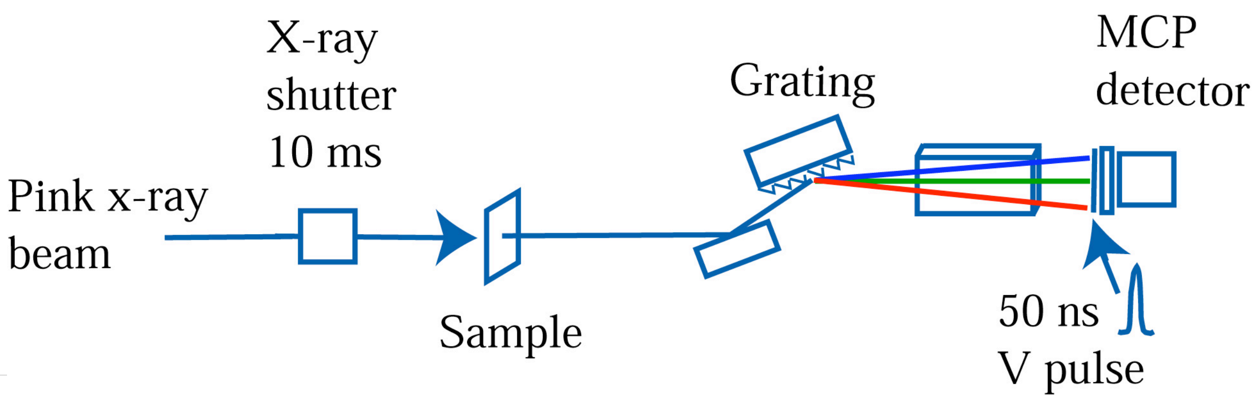
- **Plasma spectroscopy of Hot Dense Matter**
  - Use high energy laser to create uniform HED plasmas
  - Measure collision rates, redistribution rates, ionization kinetics

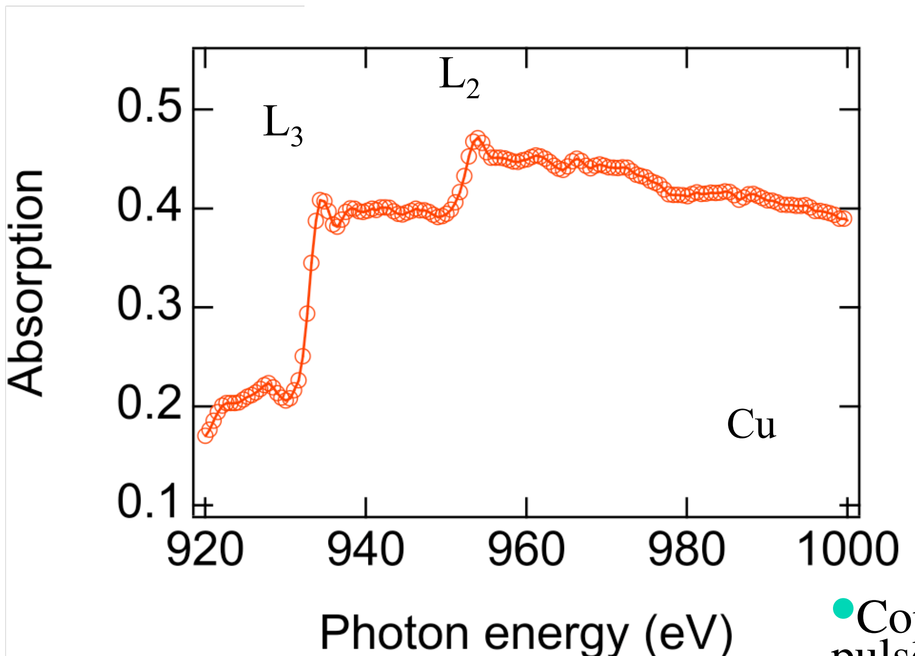


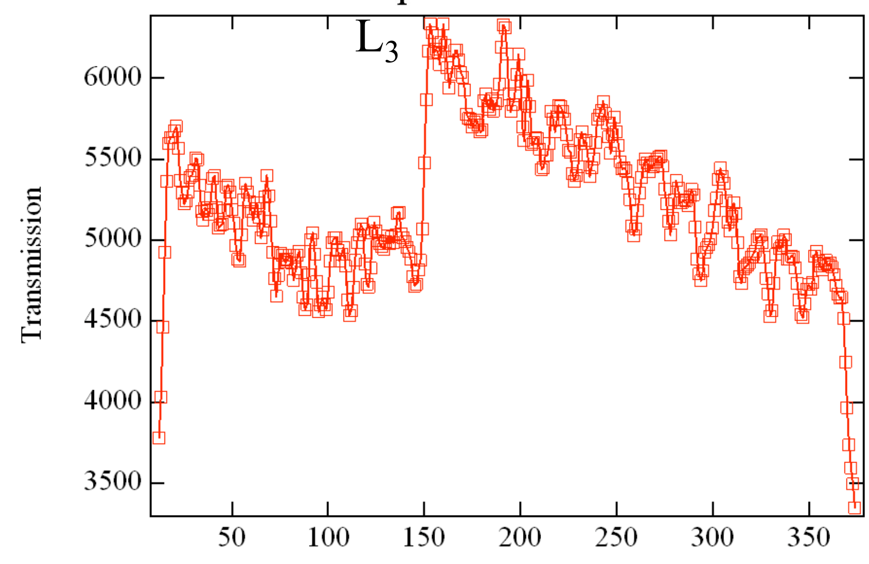
- **Probing High Pressure phenomena**
  - Use high energy laser to create steady high pressures
  - Produce shocks *and* shockless high pressure systems
  - Study high pressure matter on time scales < 1 ps
  - Diagnostics: Diffraction, SAXS, Diffuse scattering, Thomson scattering



Preparation for foils experiments: Dispersed Cu spectrum





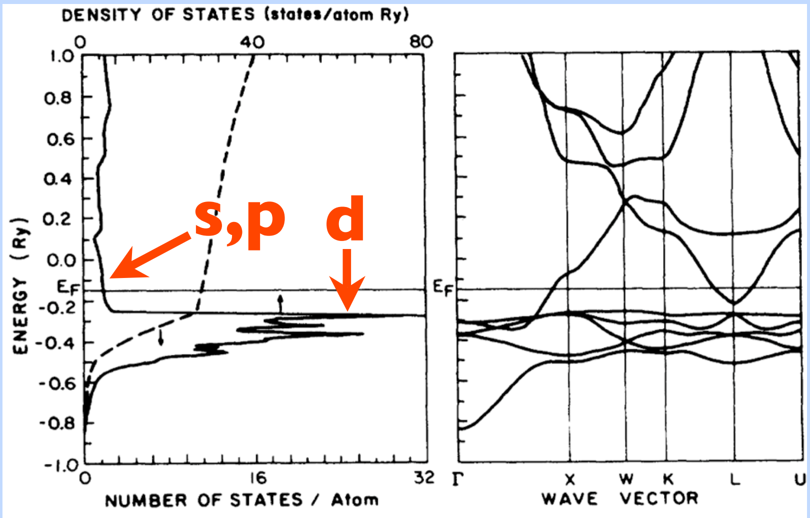


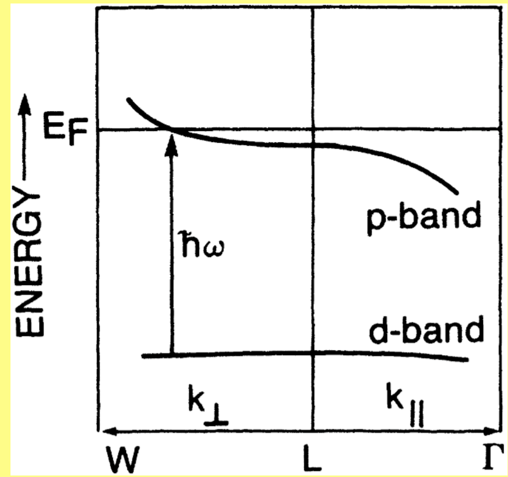
- Copper absorption spectrum measured in 10 ms

- Copper absorption spectrum measured in 10 ALS pulses
  - Intensity limited by undulator gap, aperturing beam and detector efficiency (100)

Properties of Copper

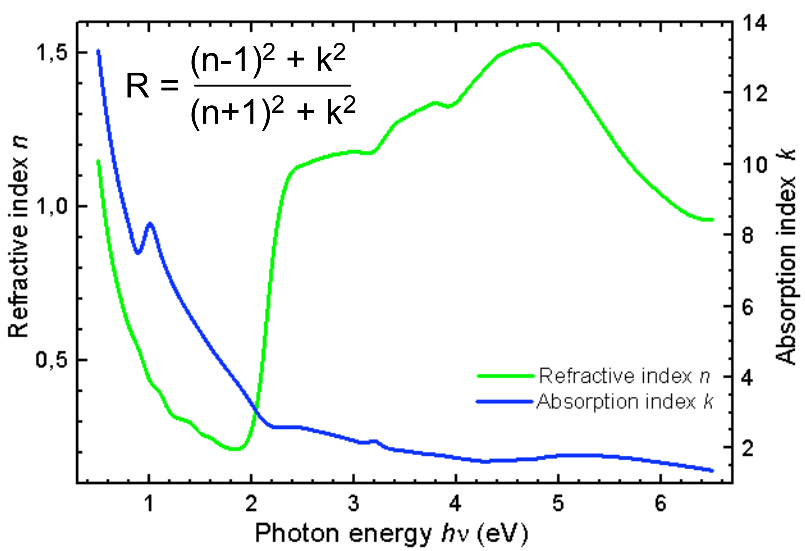
Band structure: Levy, *PRB* 1987





Optical excitation process:
Eesley, *PRB* 1986

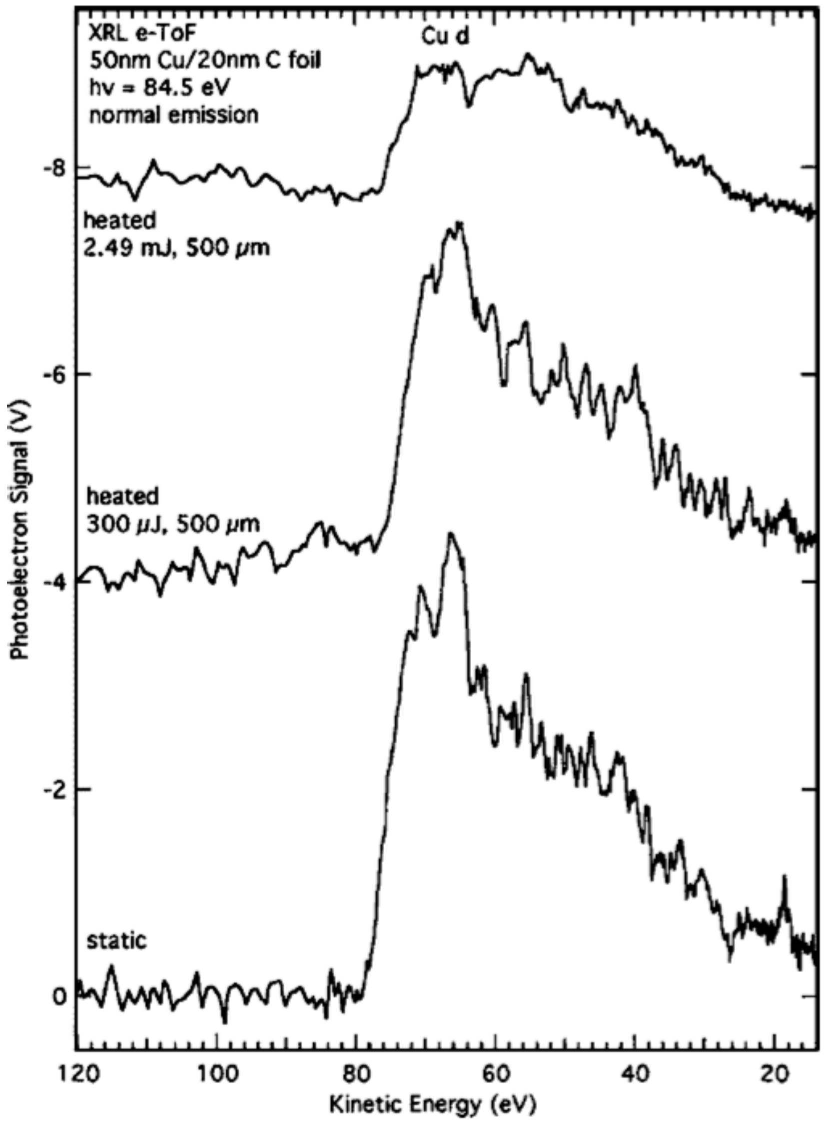
We photoexcite sample with 3eV photons. Absorption from the d-band to p-states above E_F is strong. This is the same absorption process that gives Cu its color.



K: thermal conductivity[b] (W/m K)	401
C_l: lattice heat capacity[b] (J/m^3 K)	3.43×10^6
A: electronic heat capacity[b] (J/m^3 K^2)	96.6
$\tau_{e\text{-}ph}$: electron-phonon collision time[c] (sec)	2.4×10^{-14}
α: absorptivity[d] (m^{-1})	7.1×10^7
G: electron-phonon coupling [Eq. (4)] (W/m^3 K)	2.6×10^{17}
m: electron mass[b] (kg)	9.1×10^{-31}
N: conduction-electron density[e] (m^{-3})	8.4×10^{28}
v: longitudinal sound velocity[b] (m/sec)	5010
T_d: Debye temperature[b] (K)	343
D: Debye integral[f] [Eq. (5)]	0.62

Eesley, *PRB* 1986

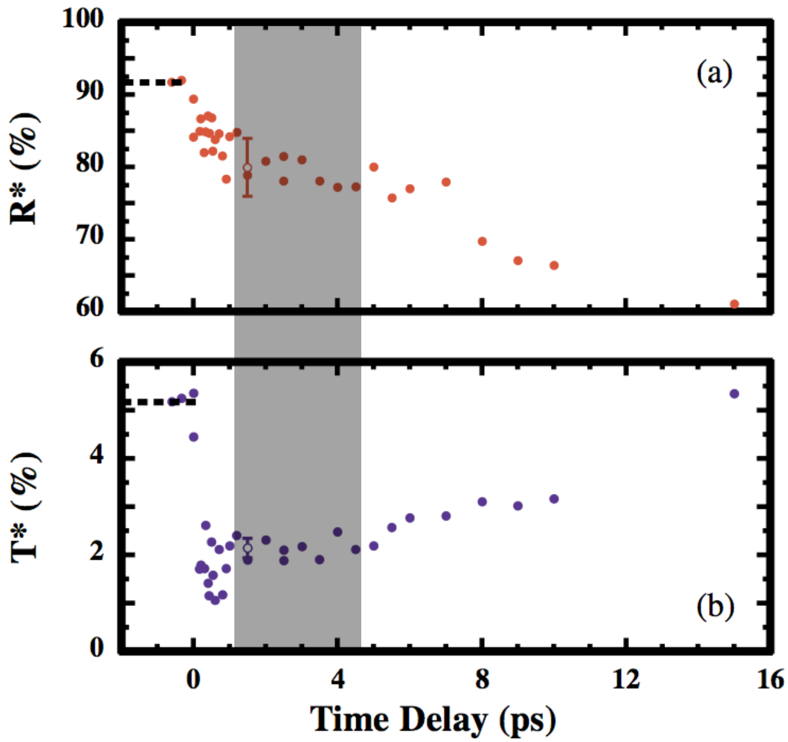
Photoemission from laser-heated Copper



Nelson, APL 2005

- Spectra measured at 0 ps < Δt < *4 ps*
- The middle curve is “heated” (300 μJ); the upper curve is ionized (2.5 mJ).
- Spot size 500 x 700 μm
- Shows rapid depopulation of the d-band

Relevant time-scale is measurable



Widmann, *PRL* 2004: transient measurements on melted gold. Found a "quasi-steady state" that lasts a few picoseconds, before the sample starts expanding; the ion cores are still comparatively cool, and the electrons are very hot. It appears that the duration of the QSS is set by electron-phonon coupling in this non-equilibrium state.

So by the time the electrons equilibrate with the lattice, the material's already expanding. So you never measure a "thermal" sample.

Elsayed-Ali *PRL* 1987 (Copper):
e-ph relaxation ~ 1 - 4 ps

Eesley *PRB* 1986 (Copper):
e-ph relaxation < 1 ps

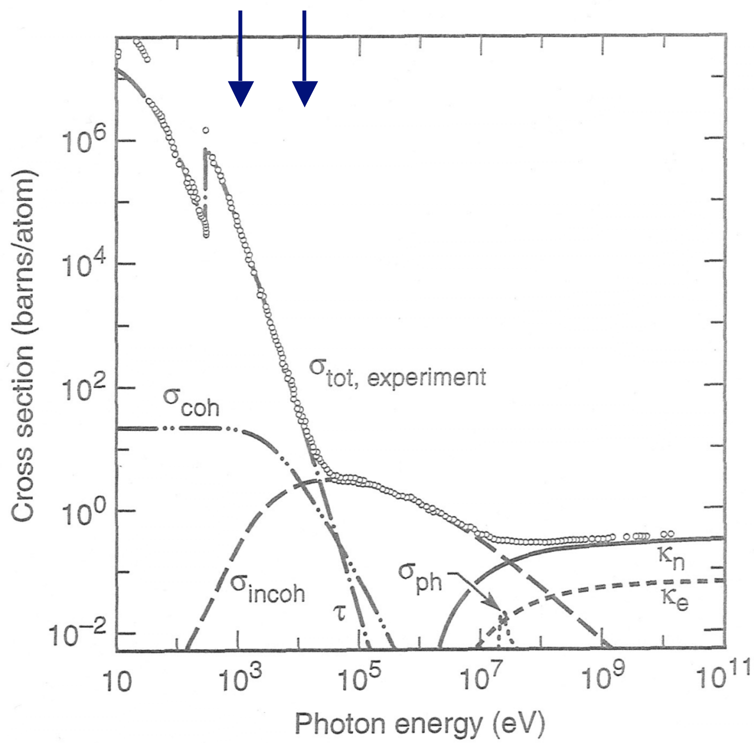
Schoenlein *PRL* 1987 (Gold):
e-ph relaxation ~ 2 - 3 ps

Widmann *PRL* 2004 (Gold):
e-ph relaxation > 5 ps

Streak camera resolution ~ 2 ps

Challenges to theorists: Absorption and scattering cross-sections

LCLS and Future
Soft X-Ray FELS



At the highest intensities (i.e., up to requirements for atomic-resolution single macromolecule imaging: 10^{22} W/cm^2):

- Does the **ratio of absorption to scattering** stay the same, affecting singe macromolecular imaging studies (dependence of damage and signal)?

- Will **Raman processes** allow useful broadbanding of the LCLS pulse, for absorption spectroscopy (NEXAFS, etc.)?

- Will **transparency or guiding** effects be important, for deeper penetration in HEDS studies?

Challenges under "warm" conditions in condensed matter, materials physics, and plasma physics can be addressed

- understand the dynamic interplay between **electronic structure** (energy levels, charge distributions, bonding, spin) and **atomic structure** (coordination, bond distances, arrangements)

Fundamental time scales range from picoseconds (conformational relaxations in molecular systems, and electron-lattice energy transfer times in solids), to ~100 fs (vibrational periods), to ~10 fs (electron-electron scattering), to <1 fs (electron-electron correlations)

X-rays are ideal probes of atomic structure, electronic structure, and plasma properties

New x-ray sources should enable the application of x-ray spectroscopic and scattering techniques (XANES, EXAFS, XMLD, XMCD, RIXS) on fundamental time-scales.